Aalto University
School of Chemical
Technology

Aalto University
School of Chemical
Technology

School of Chemical Technology
Degree Programme of Bioproduct Technology

Suchart Setasith

Effect of compressive and abrasive refining on structural changes in fiber and paper

Master's thesis for the degree of Master of Science in Technology submitted for inspection, Espoo, 14 May, 2014.

Supervisor Professor Jouni Paltakari
Instructor Doctor Eero Hiltunen

Aalto University
School of Chemical Technology

Aalto University, P.O. BOX 11000, 00076 AALTO
www.aalto.fi

Abstract of master's thesis

Author Suchart Setasith

Title of thesis Effect of compressive and abrasive refining on structural changes in fiber and paper properties

Department Bioproduct Technology

Professorship Paper technology **Code of professorship** PUU-21/P3003

Thesis supervisor Professor Jouni Paltakari

Thesis advisor(s) / Thesis examiner(s) Doctor Eero Hiltunen

Date 14.05.2014 **Number of pages** 81 **Language** English

Abstract

The objective of this master's thesis is to explain the differences in refining action and refining results between compressive and fibrillating refining types. Another aim of this work is to find out the relationship between structural changes in fiber and paper properties.

Softwood pulp samples were refined by Voith LR40, Lampén mill and MASUKO Super Masscolloider. Then fiber structural changes resulting from refining were investigated. External fibrillation, fiber curl and fines were evaluated by Kajaani fiber analyzer apparatus. Internal fibrillation was evaluated according to the degree of Fiber saturation point (FSP). Other fiber and pulp properties were also studied including water retention vale (WRV), Schopper-Riegler (SR), and Zero-span breaking strength. After that effect of refining action on structural changes was determined. Paper properties were then examined which consist of strength, optical, structural and surface properties. Finally, the relationship between structural changes in fiber and paper properties was analyzed.

The study found that Lampén refining, representing compressive refining, highly promote external fibrillation, internal fibrillation, fiber straightening and somewhat shorten fibers. On the other hand, Voith refining, representing combination of abrasive and compressive refining, was found to highly promote external fibrillation, fiber straightening but moderately fiber shortening and develop internal fibrillation. Furthermore, internal fibrillation was found to largely promote most of paper strength properties and have a moderate impact on surface and structural properties while external fibrillation and fine greatly affect structural and surface properties.

Keywords Compressive refining, abrasive refining, structural changes in fiber, internal fibrillation, external fibrillation, Lampén mill, Voith LR40, MASUKO Super Masscolloider, fiber properties, paper properties.

Acknowledgement

This thesis was carried out in Laboratory of Paper and Printing Technology at Aalto University during 2013-2014.

I would like to express my gratitude to my supervisor, Professor Jouni Paltakari and my instructor, Doctor Eero Hiltunen for his valuable comments and advices. I also thank to the all laboratory staffs, friends who help and support me throughout my work.

I would like to thank my company, SCG PAPER, which supports me for the scholarship for entire period of my study in Finland. This work could not be successful if I would not receive the supports from them.

Finally, I would like to thank to my parents and friends who always support and encourage me.

Espoo, 14 May 2014

Setasith Suchart

Contents

List of Abbreviations 3
Introduction 4
Literature Part 5
1. Principles and theories of refining 5
  1.1. Compressive and abrasive refining 5
  1.2. Effect of refining on pulp/paper properties and paper machine runnability 7
    1.2.1 Effect of refining on water retention value (WRV) 7
    1.2.2 Effect of refining on paper properties 8
    1.2.3 Effect of refining on runnability 13
  1.3. Control and characterization of refiner 15
2. Changes of fiber structure in refining 19
  2.1. Internal fibrillation and internal fibrillation measurement.................... 19
  2.2. External fibrillation and external fibrillation measurement 21
  2.3. Fiber straightening and fine formation 27
  2.4. Release of organic substances 29
3. Web forming and Dewatering.................... 29
  3.1. Fiber network and bonding.................... 29
    3.1.1 Paper structure 29
    3.1.2 Effect of fiber structure changes on inter-fiber bonds 30
    3.1.3 Relative bonded area 30
    3.1.4 Fiber segment activation.................... 30
  3.2. Water in fiber network and drainage properties.................... 31
    3.2.1 Fiber saturation point (FSP) and Water retention value (WRV) 31
    3.2.2 Canadian Standard Freeness (CSF) and Schopper-Riegler (SR).................... 31
    3.2.3 Drainage properties 32
Experiment Part 33
4. Materials and Methods.................... 33
  Materials 33
  Methods.................... 36
5. Results and Discussion 40
  5.1 Refining VS fiber structural changes 40
  5.2 Fiber structural changes VS Paper properties.................... 47

5.3 Refining type VS Paper properties 56
5.4 Summary of the results 63
6. Summary 67
7. References 68
List of Appendices 74

List of Abbreviations

CEL	Cutting edge length
CSF	Canadian Standard Freeness
FSP	Fiber saturation point
MBF	Moving Belt Former
RBA	Relative bonded area
RPM	Revolutions per minute
SEC	Specific energy consumption
SEL	Specific Edge load
SR	Schopper-Riegler
WRV	Water retention vale

Introduction

Refining plays a key role in the papermaking process to develop pulp and fiber properties by changing fiber structure, including fiber shortening, fiber straightening, external fibrillation and internal fiber fibrillation. External fibrillation refers to the peeling off of fibrils from fiber surfaces while still leaving them attached. Internal fibrillation is involved in partial delamination and pore widening within cell walls due to an externally applied load (Hartman, 1985; Kerekes, 2005; Wang, 2006, and Kang, 2007). Structural changes in fiber occur according to the type of refining. Abrasive refining mainly generates external fibrillation using a refiner which applies an abrasive force on fibers, such as a conical refiner, double disc refiner. On the other hand, compressive refining principally generates internal fibrillation using a refiner which provides compressive force on fiber, for instance Lampén refiner (Koskenhely, 2007 and Kang, 2007).

External fibrillation and internal fibrillation affect fiber and paper sheet properties differently. Both types of fibrillation largely have been found to improve various paper strength properties; however, the excessive fibrillations also causes negative effects on dewatering at forming section as well as other physical and optical properties (Page, 1989, Strachen, 1932; Mason, 1950; Emerton, 1955; Kerekes, 2005; Wang, 2006 and Kang, 2007). Therefore, it is important to understand the effect and relationship of structural changes in fiber on both dewatering and other properties. When controlling both external and internal in refining properly, it would offer optimum advantages in a fiber and a paper properties.

The aim of this thesis is to determine the effect of compressive and abrasive refining on the structural changes in fiber and paper properties. Three sets of refining techniques were used for this thesis. Compressive treatment was performed using Lampén mill refiner, while abrasive treatments were conducted using Voith laboratory refiner and Masuko Supermass Colloider. The results of Moving belt forming (MBF) were used to analyze the effect of the refining action on the dewatering.

The rest of this thesis is divided into four chapters. Chapter 2 reviews literature about principles and theories of refining, changes of fiber structure in refining as well as web forming and dewatering. Chapter 3 describes materials and methods used in this study. Results and discussion as well as summary are presented in Chapter 4 and 5.

Chapter 2. Literature Part

1. Principles and theories of refining

Refining is the approach used to improve fiber properties by mechanical force changing the fiber structure. In typical refining process, fibers are brought into the grooves of refiner bar and treated by abrasive and compressive force at the edge of the bar surface. Structural changes in fiber are affected by numerous refiner characteristics, such as refiner bar pattern, groove pattern, gap between the bar, and control parameters, including amount of refining and refining intensity. Refining internally and externally changes fiber structure fibrillating fibril on the fiber; moreover, it also releases fines and organic substances from fiber surface. External fibrillation occurs on the fiber surface, and the internal fibrillation takes place within cell wall of fiber. (Koskenhely, 2007)

Amount of refining and refining intensity can be expressed by number of impacts and severity of impacts respectively. Both number of impacts and severity of impacts affect refining results of refined fiber. Number of impacts represents the amount of the treatment. The severe impact increases fiber cutting while mild impact develops fiber swelling. The time that pulp spend on refiner (dwell time) and rotational speed (rpm) is used to determine number of impacts, while type of bar, pattern of bar, bar angle, arrangement of bar, and pressure between rotor and stator bar is correlated with the severity. Leider and Nissan proposed the mathematical equation to determine net refining energy. The equation is determined by the severity and number of impacts. Residence time of fiber and the possibility of a fiber to be in impact position are also taken into account. However, the Leider's and Nissan's equation was strongly criticized that it relies on unqualified intuition. Nevertheless, the issue on impact of cyclic refining has been still on extensive debate (Leider and Nissan, 1997; Lossada, 2001; Dekker, 2003; Kerekes, 2005; Koskenhely, 2007).

1.1. Compressive and abrasive refining

1.1.1. Compressive refining

Compressive treatment effectively generates both internal fibrillation and fiber straightening. The internal fibrillation is involved in the breakage of the crosslinks between microfibrils resulting in delamination of the fiber cell wall as shown in figure 1. Compressive treatment develops internal fibrillation in mechanical pulp less than that in chemical pulp because high lignin content in mechanical pulp restricts the development of fibrillation (Taniguchi and Okamura, 1998; Stone, Scallan, and Abrahamson, 1968). The study of Wang has shown that internal fibrillation is highly developed when refined by Lampén mill at 7,500 revolutions at pulp consistency of 3% (Wang, 2006). At high consistency, pulp is immobilized and cyclically compressed only on one side. In case of laboratory beater (PFI mill), it effectively generates internal fibrillation at higher pulp consistency, approximately around 10% consistency (Wang, 2006). Fiber straightening and internal fibrillation improve tensile strength greater than external fibrillation when considering at the same swelling degree (Murphy, 1962; Kerekes, 2005).

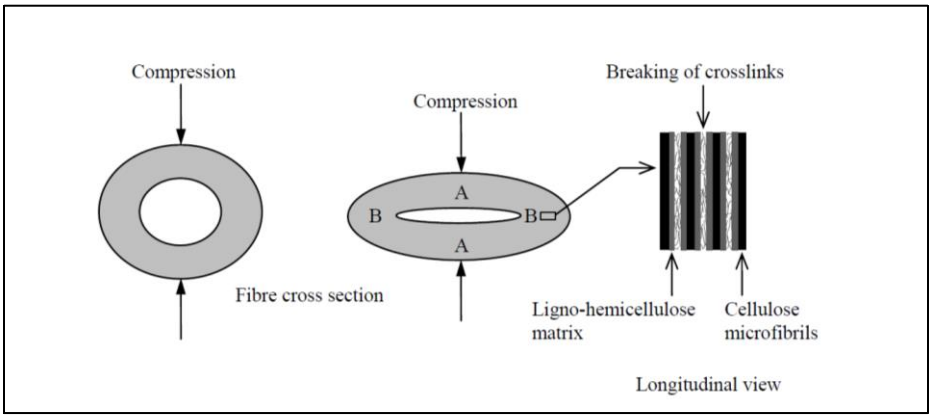


Figure 1. Generation of internal fibrillation resulting from compression force (Wang, 2006).

1.1.2. Abrasive refining

Abrasive refining is involved in abrasive action between fibers and refiner bar leading to generation of fines and external fibrillation (Wang, 2006). External fibrillation improves light scattering coefficient and Scott bond Strength (Kang, 2006) while lowering dewatering effectiveness (Kang V, 2006). The fibrillating treatment begins when pulp suspensions are brought into typical disc or conical refiner. The pulp suspension is squeezed between refining bar gap resulting in increasing its consistency. The pulp is then compressed and sheared in the gap between rotor and stator of refiner bar. These forces shear the fiber causing fines formation and external fibrillation on fiber surface (Algar and Giertz, 1951; Simpson and Mason, 1950).

External fibrillation is the after-effect of fiber swelling meaning that fiber is able to be fibrillated only after it is swollen and soft. Non-swelling fiber is rather shortened than fibrillated when it is treated because the fiber is rather stiff and brittle (Kang, 2007).

Kang's study illustrated the morphological changes in grinding process of fiber as shown in figure 2 with relatively large gap (230 μm), internal, external fibrillation, and fine formation start to form. When continuing the process further, internal fibrillation stops forming but external and fine formation still continuously increase. With the small gap (60 μm), fiber is shortened generating fines formation (Kerekes and Senger, 2006).

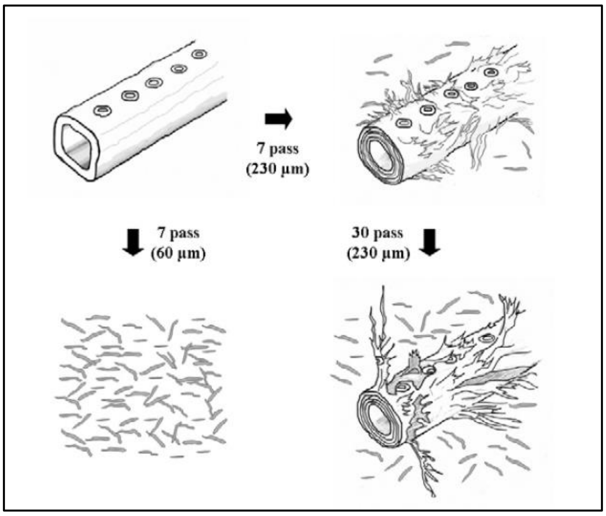


Figure 2. illustrates the morphological changes in grinding process of fiber (Kang, 2007).

1.2. Effect of refining on pulp/paper properties and runnability

1.2.1 Effect of refining on water retention value (WRV)

WRV defined as the ratio of water remaining in the pulp to dry fiber weight after centrifuging. WRV describes how tight of bonding between fiber structure and free water. It is usually used to reflect the degree of internal fibrillation or swelling property. From the figure 3, it is obvious that WRV linearly increases with refining energy meaning that refining increase ability to hold water within fiber resulting from higher swelling and internal fibrillation.

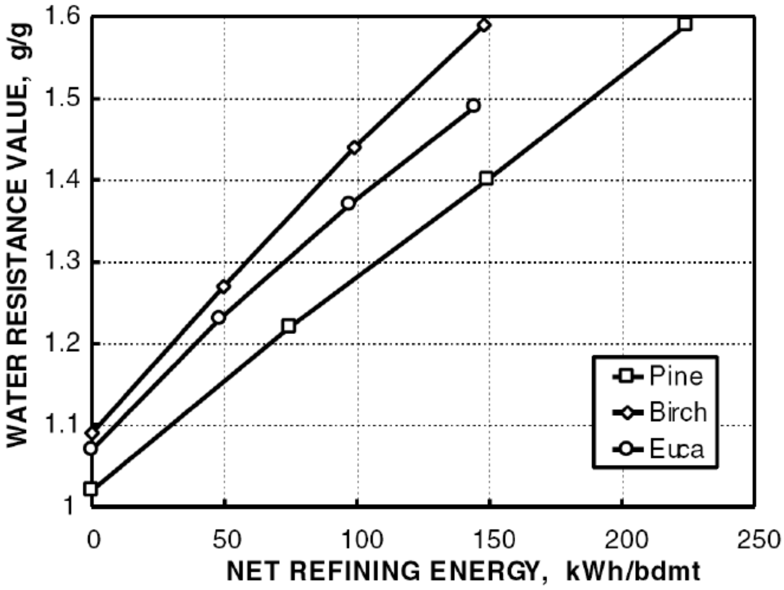


Figure 3. The effect of typical refining on WRV within three type of chemical market pulp using JC-01 refiner in serial refining with typical mill condition (Lumiainen, 2000).

1.2.2 Effect of refining on paper properties

Refining generally improves fiber bonding leading to denser paper structure. However, the responses of refining depend on type of treatment, number and severity of impact. Compressive treatment mainly provides internal fibrillation and fiber straightening while abrasive refining develops external fibrillation and fines formation. Highly severe refining shortens fiber whereas moderate refining fibrillates fiber (Lumiainen, 2000).

Structural properties

Refining generally decreases bulk as the growth of the refining because it increases fiber flexibility and reduces pore size and pore amount in the fiber network. In other words, refining causes a high density in paper structure which affects several strength properties. The result is illustrated in figure 4 (Lumiainen, 2000).

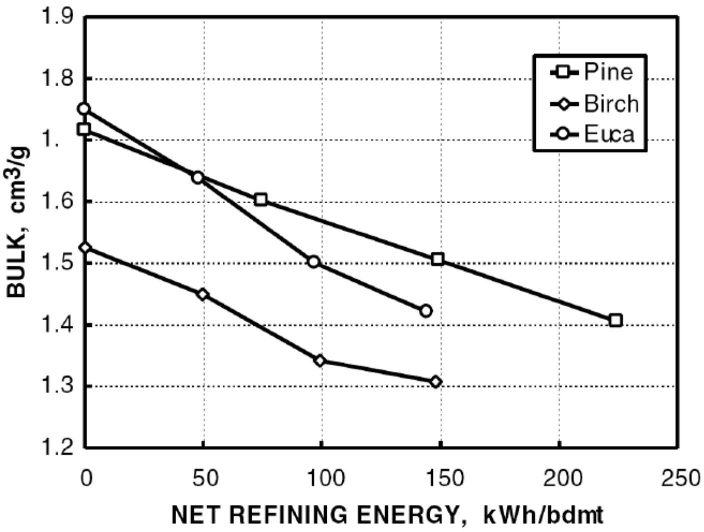
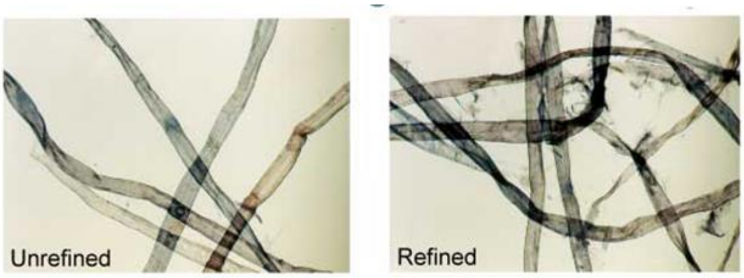


Figure 4. Effect of refining on Bulk within three type of chemical market pulp using JC-01 refiner in serial refining with typical mill condition (Lumiainen, 2000).

Strength properties

- Tensile and bursting strength

Refining basically improves fiber flexibility and increases fiber bonding area due to surface fibrillation. Moreover, fiber network activation also occur in refining resulting in increasing fiber swelling and corresponding shrinkage. Because of the higher fiber bonding competency and fiber network activation, tensile and bursting strength are increased in the same manner. Figure 5 presents that refining typically improves tensile strength index (Lumiainen, 2000).



	Unrefined	Refined (200 kWh/t)
• Freeness, ml	735	455
• Fiber length, mm	2.34	2.19
• Tensile index, Nm/g	28.4	73.8
• Tear index, mNm^2/g	20.5	16.0
• Air permeability, Bendtsen ml	1350	1040
• Bulk, cm^3/g	1.79	1.52

Figure 5. The effect of typical refining on bleached Pine fibers (KnowPap, 2000).

- Tear strength

Tear strength is affected by fiber length, individual fiber strength, as well as inter-fiber bond area and strength (friction between bonded fibers). Refining increases inter-fiber bond area resulting in higher tear strength within the beginning of the refining. However, after reaching the maximum tear strength, further refining lowers the tear resistance. Figure 6 shows impact of refining on tear strength (Lumiainen, 2000).

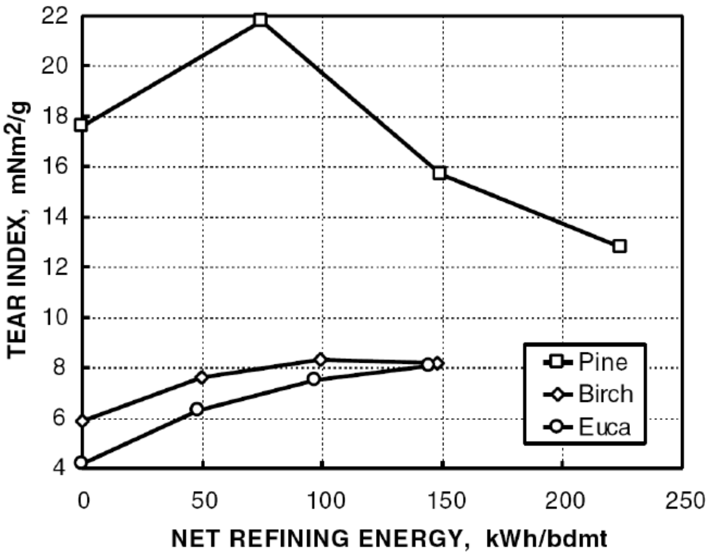


Figure 6. The effect of typical refining on tear strength within three type of chemical market pulp using JC-01 refiner in serial refining with typical mill condition (Lumiainen, 2000).

- Folding strength

Folding strength is developed by increasing fiber bonding area and strength since those denser bonds can handle more loads; however, too high amounts of fiber bond will create tensile and press stresses when folding resulting in reduction of the strength and folding failure shown in Figure 7. Therefore, properly high refining provides highest folding strength.

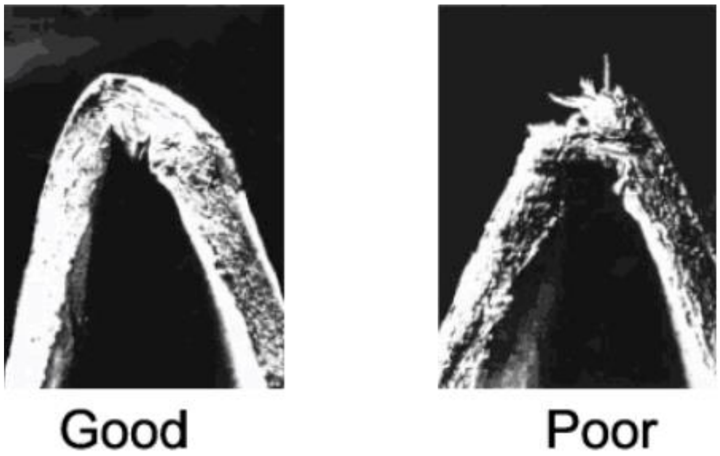


Figure 7. Left: Normal sheet condition after folding. Right: Poor sheet condition due to the failure of folding resistant (KnowPap, 2000).

- Wet strength

Wet tensile strength is one of the most widely used properties to determine runnability of paper. Lindqvist's work reported that wet tensile strength is largely increased at constant dry matter content caused by fines resulting from refining; however, other effects, such as internal and external fibrillation, are also believed to contribute to the increase of wet tensile strength. The result is presented in figure 11 (Lindqvist, 2011). However, refining typically decreases dewatering competency and dry matter content of paper thus decreasing wet strength.

Optical properties

Most optical properties rely on light scattering coefficient. Refining affects the light scattering coefficient in two opposite manner. On the one hand, in the lower specific energy consumption (SEC), refining slightly develops external fibrillation leading to expand free surface area of fibers, which increases the light scattering coefficient. On the other hand, at the higher-SEC, refining increases the amount of relative bonded area and bindings between fibers; as a result, it reduces the light scattering coefficient due to the lower amount of free surface area and pores and the differences in optical properties between fiber and air. The result is represented in figure 9 (Lumiainen, 2000).

The effects of refining on the optical properties are different according to pulp type. In chemical pulp, the negative effect of refining to the light scattering overcomes the positive effect; as a result, refining in chemical pulp lowers paper brightness property. On the other hand, in mechanical pulp, refining slightly increases brightness due to the compensation between increasing in light adsorption and higher light scattering. However, brightness is also affected by several other factors, such as PH, ion in water, and organic material (Lumiainen, 2000).

In opacity, refining develops both outer surface area and fibers binding. However, the growth of fibers binding conquers surface area growth resulting in decreasing of opacity property. The result is represented in figure 8.

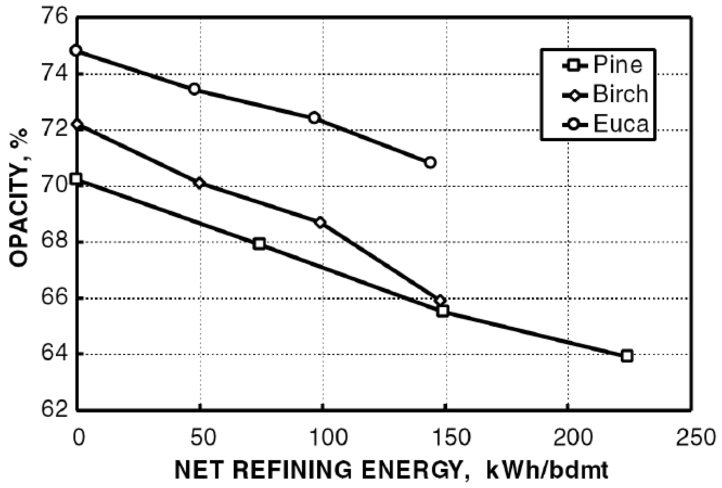


Figure 8. Effect of refining on opacity within three type of chemical market pulp using JC-01 refiner in serial refining with typical mill condition (Lumiainen, 2000).

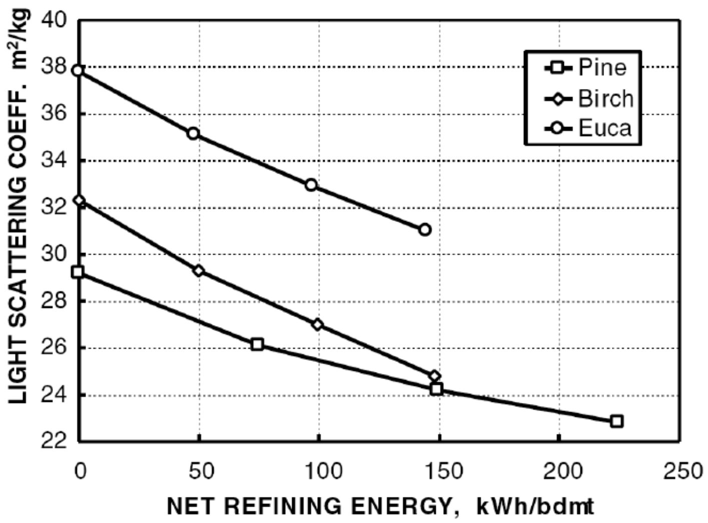


Figure 9. Effect of refining on light scattering coefficient with three type of chemical market pulp using JC-01 refiner with typical mill condition (Lumiainen, 2000).

Surface properties

Refining largely improves surface properties because it fibrillates surface fiber increasing RBA and reduces amounts of pore at the paper surface. The surface smoothness of paper increases as the refining develops according to the type of refining treatment and paper type. The more abrasive the refining is, the smoother the paper surface can reach (Gao and et. al, 2009). The result is illustrated in the figure 10.

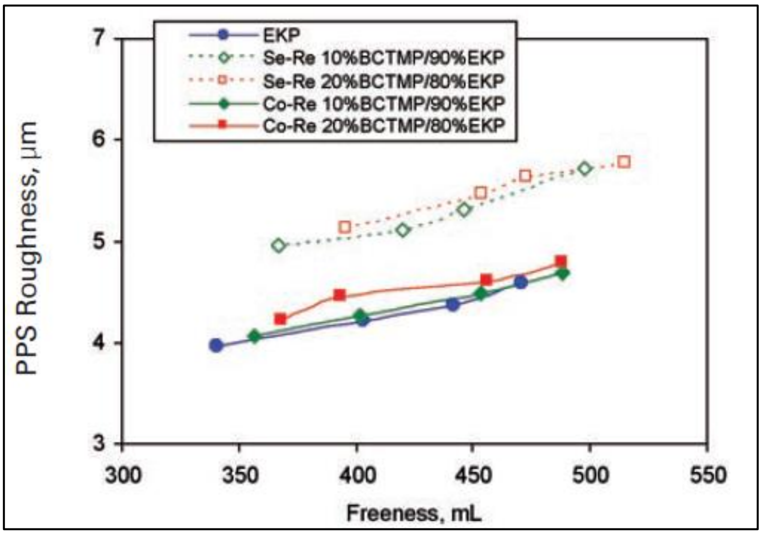


Figure 10. Effect of refining on paper smoothness of different paper (Gao and et. al, 2009).

1.2.3 Effect of refining on runnability

There are several factors affecting runnability in paper machine, including pulp and paper properties itself and its defects, process stability and pulsation, as well as efficiency of cleaning and screening in the production process. Even though stability of the process is the most significant factor affecting runnability, paper web must have high enough strength to withstand its defects and the local peak load from the process. The properties of wet web to resist the local load depend on wet strength which consists of wet and dry tensile strength, strain at break, wet fracture toughness, and dry content. The wet strength can be developed by properly increasing refining, dry content and chemical pulp content. Better dewatering improves dry content of paper web leading to increasing in paper machine runnability and in heat energy saving in drying section. Apart from mentioned properties, permeability also has an effect on sheet sealing which highly affects runnability. High permeability provides fewer problems in sheet sealing. Nevertheless, wet tensile strength is considered as one of the most important factors in runnability (Gullichsen and Paulapuro, 2000).

It has been confirmed that refining promotes both wet tensile strength when considering at the same dry solids content (shown in figure 11) and tensile strength (shown in figure 12) because fines originated from refining increase the bonded area and density of the sheet. However, the fines typically reduce permeability and dewatering on the sheet leading to a decreasing of wet runnability. The result is shown in table 1 (Lindqvist, 2011).

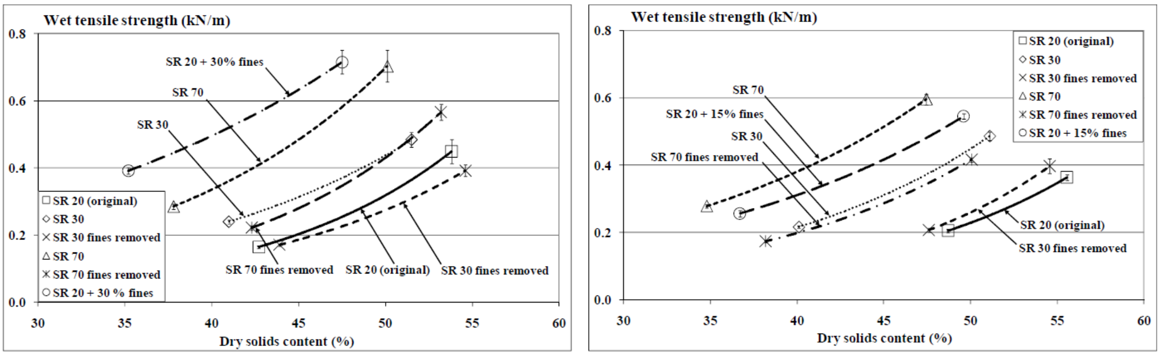


Figure 11. Wet tensile strength after refining, removal of fines or present of fine. Left: Pulp refined by beater, the sheet was pressed to two different dry contents (0.5 and 3.5 bar). Right: Pulp refined by Prolab refiner, the sheet was pressed to two different dry contents (0.5 and 1.5 bar) (Lindqvist, 2011).

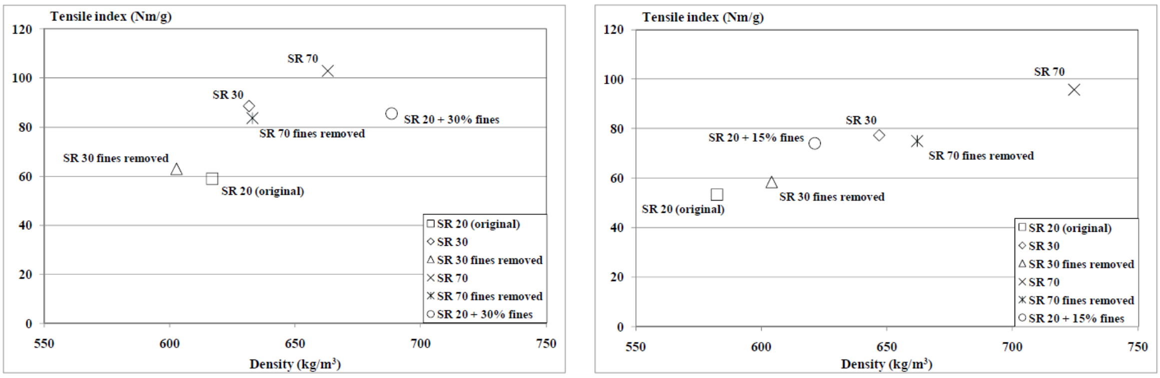


Figure 12. Tensile index versus density of dry sheet. Left: Pulp refined by the Valley beater. Right: Pulp refined by the ProLab refiner (Lindqvist, 2011).

Table 1. Dewatering time and air permeability measured with the DDÅA (Lindqvist, 2011).

	Valley		ProLab	
	Dewatering time (s)	Air permeability (bar)	Dewatering time (s)	Air permeability (bar)
SR 20 (original)	0.7 (± 0.2s)	0.31 (± 0.002)	0.3 (± 0.022)	0.25 (± 0.028)
SR 30	0.9 (± 0.1s)	0.28 (± 0.002)	0.5 (± 0.034)	0.24 (± 0.045)
SR 30 fines removed	*	*	0.2 (± 0.011)	0.27 (± 0.025)
SR 70	3.4 (± 0.1s)	0.24 (± 0.011)	5.0 (± 0.019)	0.17 (± 0.033)
SR 70 fines removed	0.6 (± 0.1s)	0.31 (± 0.016)	0.4 (± 0.048)	0.26 (± 0.029)
SR 20 + 15% fines	*	*	4.5 (± 0.095)	0.22 (± 0.064)

* = not determined

1.3. Control and characterization of refiner

1.3.1. Specific edge load and specific energy

The specific edge load or refining intensity (SEL) together with specific energy (SEC) is one of the most widely applied methods to control refining. SEL is defined as the severity of refining load, while SEC is referred as amount of the refining load. SEL can be calculated from effective refining power treating on fiber and cutting speed illustrated in Equation 1(Brecht and Siewert, 1966).

$$SEL = \frac{P_e}{L_s} \qquad (1)$$

where SEL is specific edge load [J/m or Ws/m],

P_e is effective refining power treating on fiber [kW],

L_s is cutting speed [km/s]

Cutting length refers to the total common contact length of opposite bars, number of rotor and stator bars in a filling of refiner, and rotation speed of refining. The calculation of the cutting length is shown in equation 2.

$$L_s = Z_r Z_{st} l n \qquad (2)$$

where L_s is cutting speed [km/s]

$Z_r Z_{st}$ is number of rotor and stator bars,

l is common contact length of opposite bars [km],

n is rotation speed [1/s]

The effective refining power treating on fiber is the actual power that calculated by subtracting no load power out of total power. The total power is obtained by measuring torque or power used by motor of refiner. The formula of the effective power is presented in equation 3.

$$P_e = P_t - P_{n-l} \qquad (3)$$

where P_e is the effective refining power treating on fiber [kW]

P_t is the total power [kW],

P_{n-l} is the no load power [kW].

Note that no load power is measured with water flowing through running refiner.

The amount of the refining is explained as the net specific energy consumption (SEC), which is effective refining power treating on fiber per one ton of production. Equation 4 shows the formula of SEC (Brecht, W., Athanassoulas, M., and Siewaert H. W., 1965).

$$SEC = \frac{P_e}{FC} \qquad (4)$$

Where SEC is specific energy consumption [kWh/t],

P_e is effective refining power or net power [kW],

F is flow [l/min], and

C is consistency [kg/m^3]

Even though SEL theory is commonly used to control refining in paper mill, some significant factors are ignored, such as consistency variation in the process, width of bars and grooves, the condition of filling, bar edge sharpness. Papermaker normally uses their experience to approximately determine type of filling and specific edge load to control refining. However, laboratory or pilot refiner is recommended to firstly optimize for refining of new pulp (Koskenhely, 2007).

1.3.2. Specific surface load (SSL)

Due to the fact that different kinds of fiber have distinct average fiber length, the fibers require particular treatment in order to optimize fiber refining. Moreover, specific edge load

(SEL), which is a parameter commonly used to control refining, determine only bar length but it does not consider bar width. Therefore, according to Lumiainen's study, it is necessary to take in to account specific surface load to reach optimum refining in different fibers. Specific surface load take the impact of bar width and intersecting angle (the angle in between rotor and stator when passing) into consideration as illustrated in the equation 5. IE is the same value as specific edge load (SEL) (Lumiainen, 1995).

$$SSL = \frac{IE}{IL} \qquad (5)$$

where SSL is specific surface load [Ws/m^2],

IE is energy per impact [Ws/m], and

IL is impact length [m].

The impact length can be obtained by the equation 6 as presented below.

$$IL = \frac{W_r + W_{st}}{2}\ x\ [\frac{1}{\cos\frac{\theta}{2}}] \qquad (6)$$

where W_r is width of rotor bar [m],

W_{st} is width of stator bar [Ws/m], and

θ is average intersecting angle [m].

1.3.3. C-factor

As mentioned earlier, SEL and SEC present the severity and amount of refining but do not explain how the energy impact on fiber. C-factor resolves these by taking into account the number of impacts and the energy per loading cycle per each fiber (intensity of impact) to the specific energy. The specific energy derived by C-factor is shown in equation 7 (Kerekes, 1990).

$$E = N \times I \qquad (7)$$

where E is specific energy

N is intensity of impacts

Number of impacts and intensity of impact can be calculated as equation 8 and 9

$$N = \frac{C}{F} \tag{8}$$

where N is number of impact per fiber

C is C-factor, and

F is pulp mass flow through refiner [kg/s].

$$I = \frac{P}{C} \tag{9}$$

where I is intensity of impacts

P is C-factor, and

C is net power [kW]

To determine C-factor, several parameters are taken into consideration for estimating the probability of a fiber being in the position of impact, including refiner filling geometry, pulp consistency, rotation speed, fiber length, and fiber coarseness. Nevertheless, C-factor does not consider the effect of bar edge profile, bar wearing, and bar material. The equation 10 illustrats formula of C-factor (Kerekes, 1990).

$$C = 8\pi^2 GD\rho C_F l n^3 \omega(1 + 2\tan\varphi)(R_2^3 - R_1^3)/3w(l + D) \tag{10}$$

where P is net power applied to refiner [W],

G is width of grooves [m],

D is depth of grooves [m],

ρ is density of water [kg/m^3],

C_F is pulp consistency fraction ,

l is length of fiber [m],

n is number of bars per unit arc length on stator or rotor [m-1],

ω is rotational velocity of refiner [revolution/s],

φ is bar angle from radius [degree],

R_1 is inner radius of refining zone [m],

R_2 is outer radius of refining zone [m],

w is coarseness of fiber [kg/m],

L is length of refining zone [m].

2. Changes of fiber structure in refining

Refining principally creates four types of structure changes in fiber which are internal fibrillation, external fibrillation, fiber shortening, and fiber straightening. Each structure change results in different impacts on numerous paper properties, including strength, physical, optical properties and dewatering (Murphy, 1962 and Kerekes, 2005). However, Wang's study pointed out that to reach the optimum benefits, chemical pulp refining should pay attention to increase internal fibrillation and fiber straightening while keeping external fibril and fine amount at proper level (Wang, 2006). More details about effects of fiber structure change on paper properties are discussed in this section.

2.1. Internal fibrillation and internal fibrillation measurement

Internal fibrillation is developed from the cyclic compressive load in refining. The cyclic load must overcome compression and bending strain in order to delaminate fiber cell wall. In the process of cell wall delamination, the compressive load from refining breaks intra-fiber hydrogen bond of fiber cell wall to promote internal fibrillation. Figure 13 shows cross section of internal fibrillation in fiber cell wall. The compressive load are various, including stressing, pressing, bending, flexing, curling, bruising, kneading, rubbing, twisting and crushing (Hartman, 1985). Modulus of elasticity and coarseness are the characteristics of fiber which resists to the required force to create internal fibrillation. However, some resisting characteristic are rather complicated, for instance fiber diameter; small diameter increases resisting compression strain but it decreases resisting bending strain (Koskenhely, 2007).

The internal fibrillation is involved in the breakage of the crosslinks between microfibrils and swelling in amorphous parts of the cell wall not only due to cyclic compressive loading, Kerekes added that fiber needs to continuously turn over during the compressive refining (Kerekes, 2005).

Figure 13. Cross section of internal fibrillation in a frozen hydrated sulfite fiber beaten for 120 minutes in a Lampén mill (Borch, Lyne, Mark, Habeger, ed., 2001).

Effect of internal fibrillation on paper properties

Internal fibrillation largely affects strength and optical properties of paper. Fiber becomes more flexible when its cell walls are delaminated. The more flexible fiber improves fiber straightening and fiber bonding thus increasing in tensile strength and Scott bond strength (Wang, 2006). Internal fibrillation develops Scott bond strength during refining in two steps; first, the internal fibrillation increases the low level of the strength in the beginning phase of refining (low SEC). Then, external fibrillation has higher effect in the higher external fibrillation at higher SEC (Wang, 2006). Hartman reported that internal fibrillation predominantly affects increasing of breaking length (Hartman, 1985).

It was reported that internal fibrillation improves tensile strength more than the external fibrillation. Internal fibrillation increases fiber swelling and corresponding fiber shrinkage during drying process thus increasing in fiber segment activation in fiber network leading to higher tensile strength and stiffness. However, internal fibrillation develops both light scattering and Scott bond strength lower than external fibrillation because the external fibrils from refining increase fiber surface to scatter light. In summary, internal fibrillation provides higher bulk, higher wet and dry tensile strength, as well as poorer dewatering;

while, external fibrillation provides higher Scott bond strength and light scattering coefficient, as well as lower porosity (Wang, 2006).

Methods to measure internal fibrillation

Due to the fact that internal fibrillation results from delamination of the fiber cell wall and swelling of amorphous parts of the cell wall, that correlates well with fiber pore volume (Koskenhely, 2007). The higher pore volume presents greater degree of the FSP and internal fibrillation as shown in figure 14. (Wang, 2006). The broken intrafiber hydrogen bonds can be replaced by water molecules, bound water in cell wall (Hartman, 1985). Therefore, the level of swelling impacted by refining can be determined as the degree of internal fibrillation (Koskenhely, 2007).

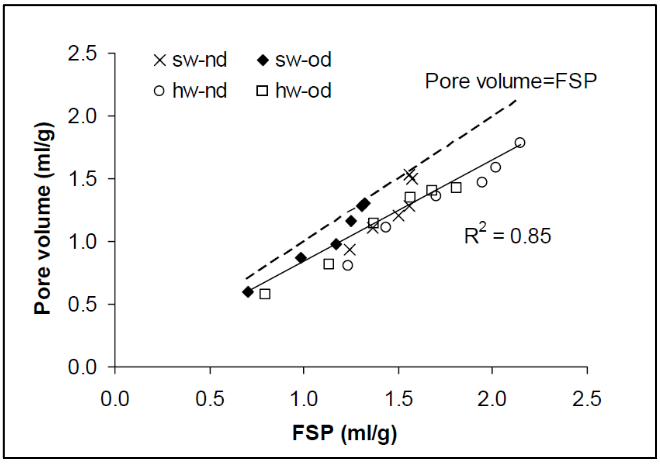


Figure 14. Relationship between pore volume and FSP (Wang, 2006).

There are many possible methods to measure internal fibrillation, related to centrifuging, solute exclusion, Nuclear magnetic resonance (NMR), inverse size exclusion chromatography (ISEC), and thermoporosimetry (Jayme, 1958; Stone and Scallan, 1968; Li, 1991; Berthold, Salmen, 1997; Maloney, Paulapuro, 2001). One of the best laboratory approaches to determine fiber swelling is to measure fiber saturated point (FSP) by the solute exclusion technique which considers only water in cell wall of fiber (Koskenhely, 2007; Stone, 1968). In many practical cases, WRV was proved to correlate well with FSP.

2.2. External fibrillation and external fibrillation measurement

The external fibrillation is developed by the peeling-off mechanism mainly resulting from abrasive action that primary wall and S1 layer are peeled off to act as bonding agents, and S2 layer is exposed to inter-fiber bond, and those fibrils still attach to the fiber surface as presented in figure 15 (Hartman, 1985; Forgacs, 1963; Corson, 1989; Sundström, 1989, Kerekes, 2005 and Kang, 2007). However, it can be also produced by non-mechanical action which is ultrasonic treatment (Kang, 2007).

Because external fibrillation changes fiber structure and increases specific surface area of fiber, therefore it somewhat affects paper properties (Cress, 1931 and Kurhila, 2005). External fibrillation is considered as the second most important factor in structural changes in fiber, after internal fibrillation, affecting web strength and it is more likely to be influenced in strength improvement in mechanical pulp than in chemical pulp (Claudio-da-Silva 1982 and Kang, 2007).

The role of external fibrillation has been controversial for over decades. Strachan and Clark presented that the external fibrillation plays a key role on the cohesion between fiber's surfaces acted as bonding agents for inter-fiber bonding (Clark, 1969 and Strachan, 1938). Many other researches reinforce that external fibrillation are largely involved in higher paper strength in paper because of the higher strength in inter-fiber bonding (Jayme and Hunger, 1957, Page and Sargent, 1961, Buchanan and Lindsay, 1961). It was reported that external fibrillation improves the retention of fine particle, including fillers, dyes and other added chemical. Even though external fibrillation is more likely to improve the retention of fine particles, it deteriorates dewatering property. (Page, 1989; Strachen, 1932; Mason, 1950 and Emerton, 1955).

Even if many studies support that external fibrillation improves paper strength, there are still several arguments claiming that external fibrillation is not involved in increasing paper strength during refining. Harrison pointed out that external fibrillation does not improve paper strength because the fibrillation develops in the stage that strength properties already remain constant. Moreover, peeling off of external fibril from fiber surface decreases individual fiber strength (Harrison, 1931 and Tasman, 1969).

External fibrillation affects light scattering coefficient oppositely between mechanical and chemical pull. Light scattering coefficient of mechanical pulp increases with refining whereas that of chemical pulp decreases (Cohen, 1948).

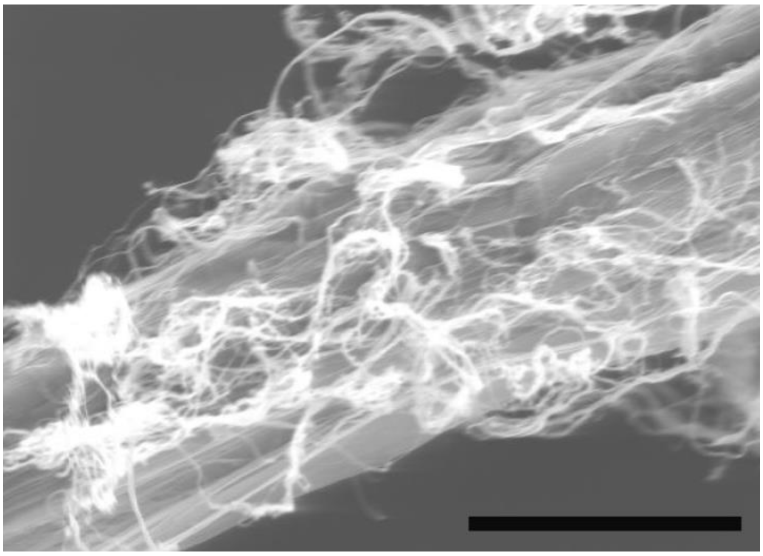


Figure 15. Image of external fibrillation of once-dried bleached kraft softwood pulp beaten for 90 minutes in a Valley beater, Scale bar of 20 µm (Kang, 2007).

Effect of external fibrillation on paper properties

External fibrillation does not significantly improve tensile strength comparing to fiber straightening and internal fibrillation according to the result of Voith-refined pulp (without fine) in figure 16. Furthermore, when determining tensile strength of pulp with and without fines (R200), fines notably reinforces tensile strength in low straightening pulp as illustrated in figure 16 (Kang, 2007).

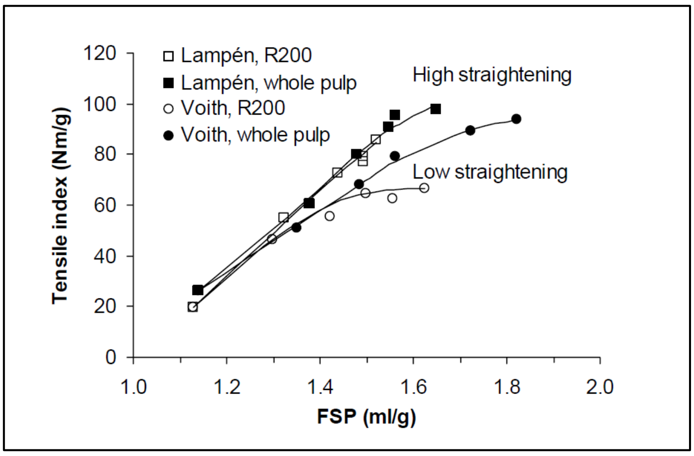


Figure 16. Tensile index versus FSP (Kang, 2007).

In optical properties, refining provide the different result of light scattering property between chemical pulp and mechanical pulp. The refining of chemical pulp generally reduces the light scattering coefficient of paper whereas that of mechanical pulp increases the light scattering coefficient. Regardless of pulp type, external fibrillation affects light scattering in two ways. Firstly, it expands fiber surface area which will increase light scattering coefficient. However, it simultaneously increases amount of bindings between fibers which then reducing light scattering coefficient as shown in figure 17 (Wang, 2006).

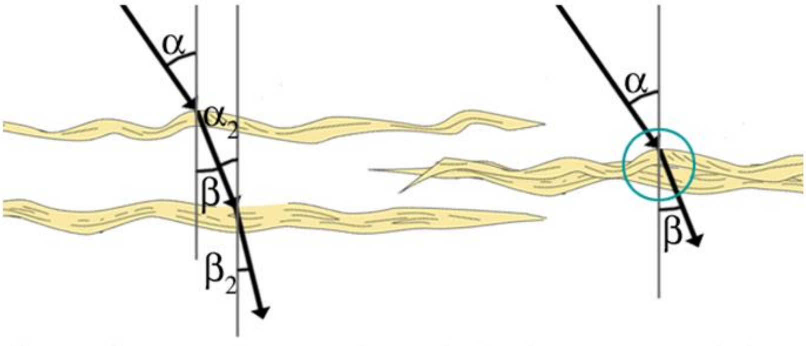


Figure 17. The light is scattered differently in layer of fiber-air-fiber in the left picture from fiber-fiber binding in the right picture (knowpap, 2000).

Besides strength and optical properties, the external fibrillation improves the retention of fine particle, including fillers, dyes and other additives, on fiber web as illustrated in figure 18 and figure 19; however, it insignificantly affects drainage properties of the pulp as shown in figure 20 (Mason, 1950; Emerton,1955 and Kang, 2007).

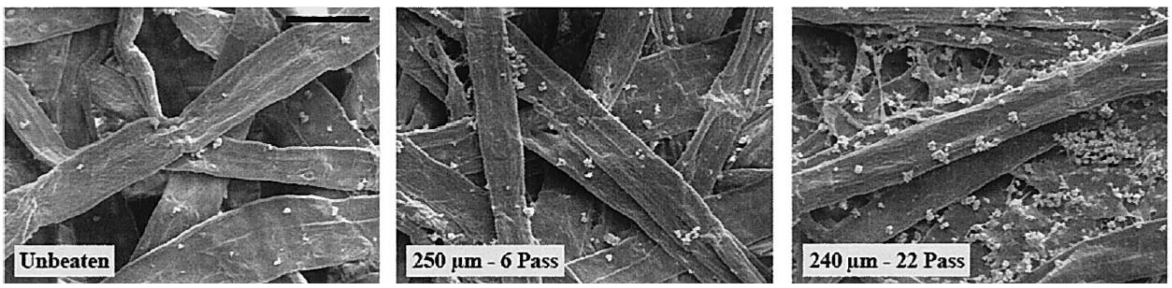


Figure 18. SEM image of filler retained in the fiber network with 30% of filler addition. The results were measured with Scale bar of 50 µm (Kang, 2007).

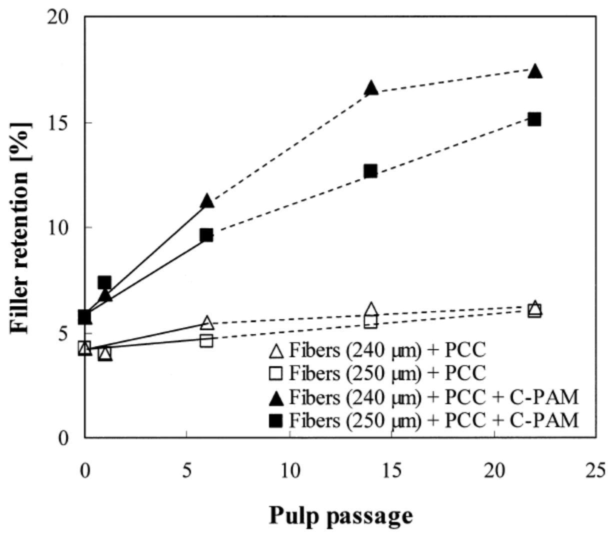


Figure 19. Effect of external fibrillation on the retention of filler (Kang, 2007). The degree of fibrillation is characterized by the amount of pulp passage (Kang, 2007).

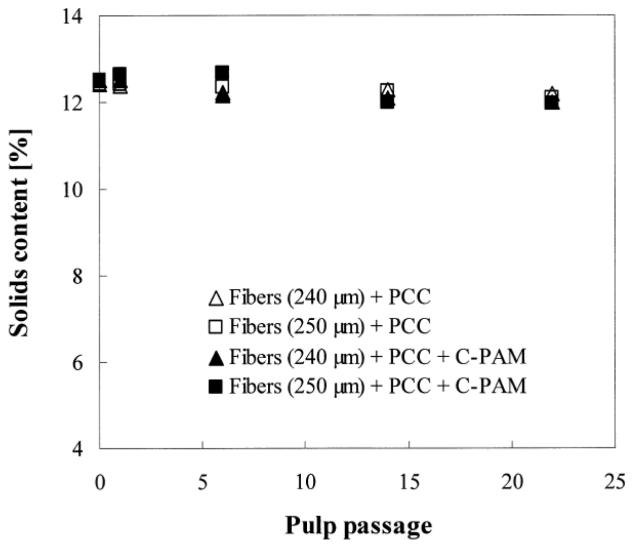


Figure 20. Effect of external fibrillation on solids content of wet sheet containing filler (The degree of dewatering property is characterized by solids content of wet sheet) (Kang, 2007).

Controversy on strength improvement by external fibrillation

As briefly mentioned earlier, even though several studies show that external fibrillation increase numerous paper properties, some studies argued that external fibrillation does not improve paper strength because the external fibrillation is generally developed at the higher SEC after the paper strength remains constant. (Harrison, 1931 and Kang, 2007) Moreover, it reduces tensile strength due to the loss of fiber strength resulting from peeling-off mechanism (Tasman, 1969 and Kang, 2007). Gallary and Cottrall added that the external fibrillation weakens the inter-fiber bond even though it increases bonded area (Gallary, 1949 and Cottrall, 1950).

Effect of external fibrillation on dewatering

The external fibrillation decreases dewatering of pulp since the fibrils are more swollen and be able to hold more water than fiber (Kang, 2007). In figure 21, Wang also supports that both external fibrils and fines significantly decrease moisture content after wet pressing comparing to internal fibrils (Wang, 2006). This means that the external fibrils and fines affect water transport within inter-fiber pores which play an important role in press dewatering. The result shows in figure 21.

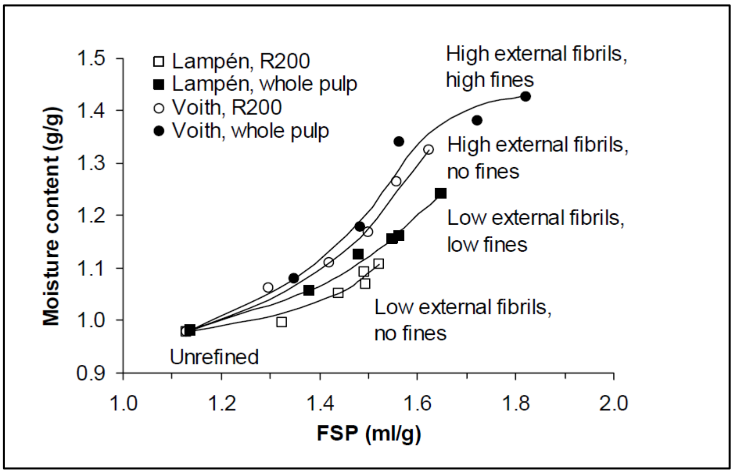


Figure 21. Moisture content after wet pressing versus FSP for fine-free pulps and whole pulps (Wang, 2006).

Methods to measure changes in external fibrillation

The methods to measure external fibrillation can be classified into two groups which are specific surface measurement and microscopy method (Kurhila, 2005). Specific surface measurement has several commonly used approaches, including gas adsorption, liquid permeability, liquid filtration, sedimentation, silvering method. However, liquid permeability method is the most widely used method for specific surface measurement. The other mentioned methods are unreliable and laborious; consequently, the result of the measurements can be interfered by other effects beside external fibrillation (Kurhila,2005).

Numerous microscopy techniques are used to determine external fibrillation, such as light microscopy, confocal laser scanning microscopy (CLSM), contrast microscopy, atomic force microscopy (AFM), scanning electron microscopy (SEM) (Weise and Paulapuro, 1996; Kurhila, 2005). Disadvantage of SEM is that fibers need to be dried; as a result, the drying can induce changes in fiber surface. E-SEM is then developed to improve SEM technique so that fiber is allowed to have certain moisture for measuring. AFM can be used solely for limited area so that small amount of fiber can be determined in each measurement. Even though light microscopy is commonly used, it is laborious approach. Both light microscopy and CLSM can proceeded further to image analysis in order to quantify external fibrillation, but CLSM provides a better resolution of image. However, these two methods are laborious because fibers need to be stained before measuring (Ora, 1987). Phase contrast microscopy seems to be a better alternative because fiber sample does not need to be stained; moreover, the resolution of its result is sufficient to observe external fibrillation (Kurhila, 2005). Apart from specific surface measurement and microscopy measurement, Wang's study shows that freeness of pulp which its fines is removed correlates well with the degree of external fibrillation (Wang, 2006).

2.3. Fiber straightening and fine formation

2.3.1. Fiber straightening

Fiber straightening can happen not only in refining process but it also takes place during the process of mixing or pumping in papermaking. Fiber curl in chemical pulp is likely to occur permanently in kraft cooking and bleaching process. The rest can be recovered by refining. Fiber tends to be more straightened when refined at low consistency. However, the refining parameters to straighten fiber are not fully understood (Koskenhely, 2007).

The earlier studies have shown that increase in fiber swelling and tension can improve fiber strengthening. However, recently, Wang has found that compressive force in refining can effectively develop fiber straightening (Wang, 2006). Straightness of fiber is typically determined by curl index. Curl index of 10% is considered as straight fiber, while that of 20% is represented curly fiber (Jayme and Hunger, 1962). Figure 22 illustrates the effect of four types of refiner on fiber curl which refers to fiber straightening and Lampén refiner obviously provides most effective fiber straightening.

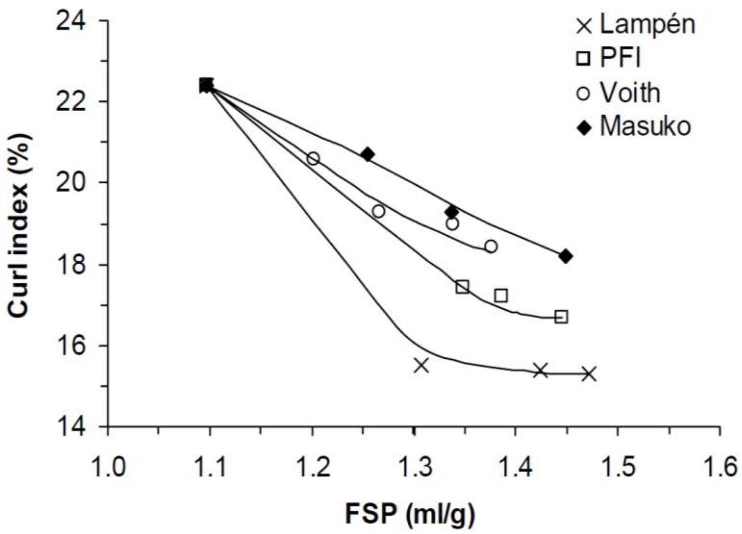


Figure 22. Effect of four type of refiner on fiber curl index against FSP (Wang, 2006).

Effect of fiber straightening on paper properties

Straightened Fiber improves tensile strength as a result of the improvement of load transferring efficiency of the fiber (Page, 1985). In addition, the study of Seth and Steenberg has supported that straightened fiber treated by both wet pressing and refining provides higher tensile strength than curled fiber (Seth, 2003).

2.3.2. Fiber shortening (fine formation)

Fiber shortening occurs when a fiber is broken or deformed due to critically high strain built up on the fiber by abrasive force from refining (Hartman, 1985). Fiber particle is defined as a fine when it is able to pass through 75 μm-diameter round hole or a 200-mesh screen of a fiber length classifier. There are two types of fine which are primary and secondary fines: primary fine refers to tiny elements originally present in wood such as shortened fibers, vessel elements and ray and parenchyma, secondary fine are fiber which is shortened during refining process (Seth, 2003).

Fiber shortening takes place due to tensile failure in fiber. The degree of fiber shortening in low-consistency refining is affected by two major parameters which are refiner load and bar edge condition. Increase of refiner load decreases the refiner plate gap, and as a result fiber experiences more axial tensile strain. If the fiber strains over its limitation, then it will be shortened. Besides effect of refiner load, too sharp bar edge and scratched edge can also cause fiber shortening. Furthermore, filling material of refiner should be properly selected according to fiber type in order to avoid too severe fiber shortening (Koskenhely, 2007 and Seth, 2003).

Effect of fiber shortening on paper properties

Higher beating level produces a large amount of fines resulting in lower freeness because fine swells and carries more water than fiber. Moreover, fines cause higher density and shrinkage in paper web. Fines increase paper density and tend to improve strength properties, including tensile strength, elastic modulus, zero-span tensile strength and double fold; on the other hand, light scattering coefficient and air permeability significantly decrease. Therefore, it can conclude that fines significantly improve fiber network bonding but it decreases dewatering efficiency (Seth, 2003; Lindqvist et al, 2011).

2.4. Release of organic substances

Refining also discharges organic substances which are carbohydrates and lignocellulose from fiber into its surrounding water. The degree of discharge reaches the highest level at the beginning of refining and gradually decreases down. Bleached hardwood pulp releases more organic substances than bleached softwood pulp; moreover, the molecular weight of the organic substances from hardwood is greater than that of softwood. Salt concentration and charge in pulp affect the releasing of organic substances. Higher salt concentration moderates the releasing of the organic substances, and higher charge density encourages the release (Koskenhely, 2007).

3. Web forming and Dewatering

3.1. Fiber network and bonding

3.1.1 Paper structure

Paper web has largely random distribution of fiber network in three-dimensional structure apart from fiber orientation. However, since fiber network's thickness is way shorter than fiber length, two-dimensional structure largely influences paper properties as well. In two-dimensional network geometry, degree of connectivity and cohesion among fibers in two-dimensional network has largely impact on mechanical properties of paper. The degree of connectivity and cohesion is characterized by relative bonded area (RBA). RBA is defined as bonded surface area of fibers divided by their total surface area. On the other hand, paper is three-dimension structure. Pores in paper network play a significant role in three-dimension relative bonded area (Niskanen, Kajanto and Pakarinen, 2000).

3.1.2 Effect of fiber structure changes on inter-fiber bonds

Inter-fiber bonds gradually increase during web dewatering as a result of surface tension forces, such as colloidal interaction pulling fibers close together. Those interactions between adjacent fibers can be reinforced by external fibrils and fines. External fibril helps bond two fiber surfaces more closely; in addition, fines improve sheet consolidation as a result of increasing of relative bonded area. The inter-fiber bonds can be characterized by wet tensile strength. Fiber swelling and shrinkage, on the other hand, is largely affected by internal fibril and chemical composition of the fiber wall. Shrinkage is directly proportional to degree of swelling of wet fiber wall which is a result of internal fibrillation. The internal fibrillation causes fiber swelling due to the fact that it increases fiber flexibility (Retulainen et al., 1998).

3.1.3 Relative bonded area

As mentioned previously, relative bonded area is determined by bonded surface area of fibers divided by their total surface area. Degree of RBA increases with higher basis weight of fiber network in two-dimensional structure while it decreases with higher number of pores in three-dimensional structure. Moreover, in practice, RBA is also influenced by pulp type, beating level, and wet pressing. The direct methods to measure RBA are quite laborious and takes a lot of time for instance nitrogen gas adsorption. Therefore some other indirect methods are usually used to measure RBA; one of the most widely used methods is Kubelka-Munk light scattering coefficient. However, this method is not highly reliable because of the fact that fiber surface will be taken into account bonded when the distance between two fibers is shorter than half wavelength of light (Niskanen, Kajanto and Pakarinen, 2000).

3.1.4 Fiber segment activation

Activation is the recovery process of deformed fiber segment in fiber network to be able to carry load. The process takes place during drying when lateral shrinkage of fiber converts into axial shrinkage of adjoining fibers at bonded area. Activation degree largely depends on inter-fiber bonding and shrinkage of fibers. Refining or beating can enhance activation due to increasing in the relative bonded area (RBA) and fiber flexibility through delamination. Several researches have reported that activation reinforces tensile strength of the sheet. Figure 23 illustrate activation process which deformed fiber segments are modified to active components of network. (Eero, 2003 and Vainio, 2007)

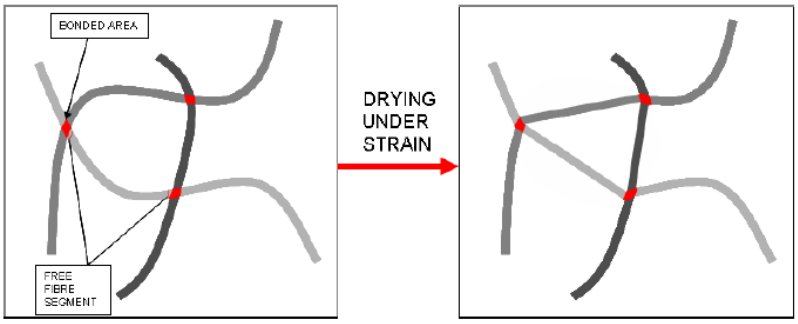


Figure 23. Schematic illustration of activation.

3.2. Water in fiber network and drainage properties

3.2.1 Fiber saturation point (FSP) and Water retention value (WRV)

Fiber saturation point (FSP) refers to the point at which all free water has been removed from the cell pore and cavities of fiber and only bound water remains in the cell walls. FSP, measured by solute exclusion technique, is one of the best laboratory approaches used to determine fiber swelling (Wang, 2006). Water retention value (WRV) is defined as the ratio of water remaining in the pulp to dry fiber weight after centrifuging. WRV describes how tight of bonding between fiber structure and free water; moreover, it increases with increasing in beating as a result of the growth of internal fibrillation and external fibrillation, such as widening pore and delamination of fiber wall. WRV yields the result of fiber development in refining and water removal at press section (Kajanto, Niskanen, 2000 and SCAN-C 62:00, 2000).

3.2.2 Canadian Standard Freeness (CSF) and Schopper-Riegler (SR)

Both CSF and SR are the most commonly used to measure drainability of a pulp suspension. However, they do not explain the potential of pulp drainage on paper machine. CSF and SR are rather used to describe the refining consequence in chemical pulp during beating and fiber coarseness in mechanical pulp after refining or grinding. These two methods have the same principle but the method to indicate drainability describes in opposite meaning. SR number is directly proportional to the drainage resistance whereas CSF number is inversely proportional. In the other word, the smaller number of CSF means better drainage resistance. The measure result is affected by several factors, such as fines content and water quality, i.e., PH, electrolyte content, and hardness. The figure 24 shows CSF apparatus (Hiltunen, 2000).

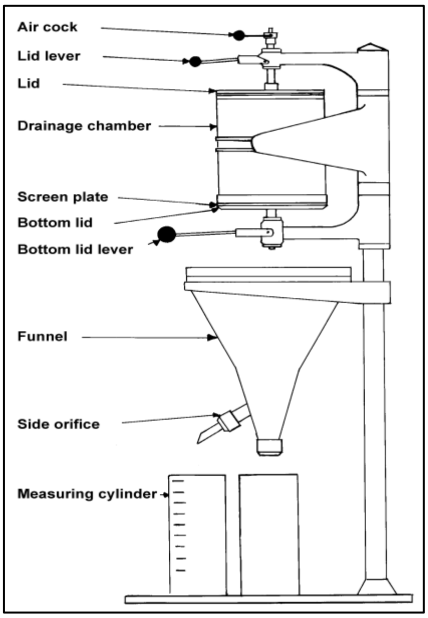


Figure 24. CSF apparatus (Hiltunen, 2000).

3.2.3 Drainage properties

Drainage is one of the most significant parameters used to evaluate the capability of dewatering at the wet end in papermaking process. Drainage behavior of pulp suspension is usually characterized by Dynamic Drainage Analyzer (DDA) shown in figure 25 or Moving Belt Former (MBF) which has been developed for simulating the wire section of a paper machine (Karrila, Raisanen and Paulapuro, 1992). MBF and DDA provides more reasonable of pulp drainage property than CSF and SR. Apart from amount of water removal, drainage time is another important parameter which is the time measured during water removal in sheet formation process. It greatly depends on pressure difference generated by drainage, density of fiber mat on the wire, fiber properties, such as fiber length, flexibility and swelling (Strengell, et al., 2004 and Blomstedt, et al., 2010).

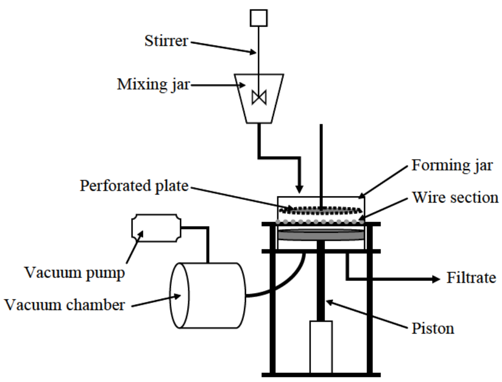


Figure 25 The Dynamic Drainage Analyzer, DDÅA (Lindqvist, 2011).

Experiment Part

Chapter 3. Materials and Methods

Materials

Pulps

A bleached softwood pulp produced in a Finnish pulp mill was refined. The pulp had a dry content of about 95.0%. The SWK was a mixture of Scots pine and Norway spruce from Finland. The pulp was stored at room temperature.

Refiner

- Lampén mill

The Lampén mill is a rotating-ball mill with a 10-kg ball and housing rotating around a horizontal axis at 250 rpm. The figure 26 shows Lampén mill refiner.

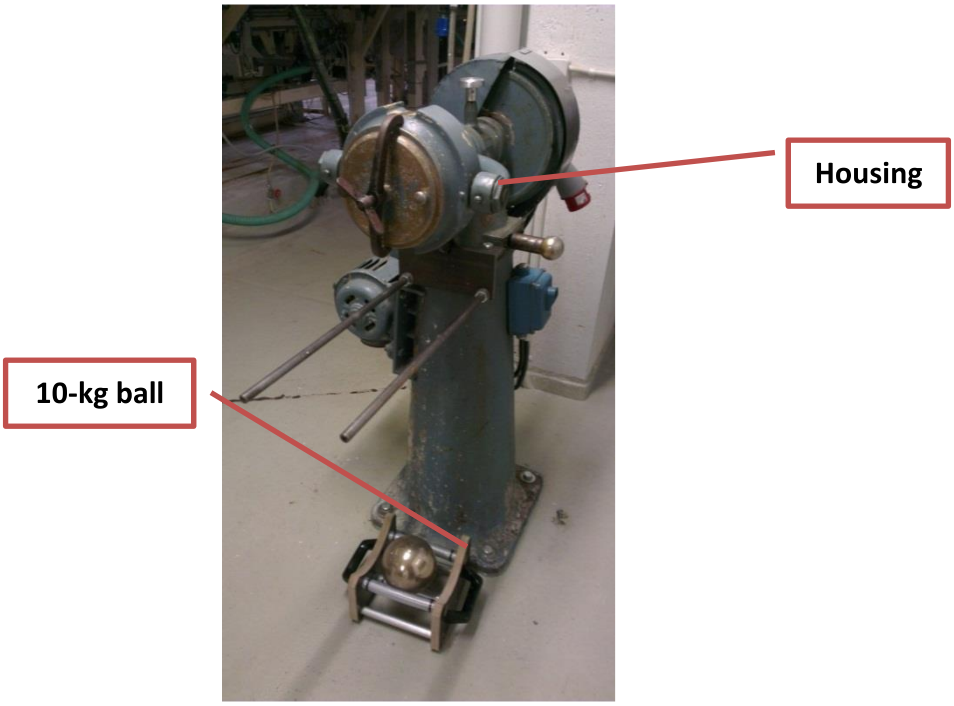


Figure 26. Lampén mill refiner at PUU laboratory, Aalto University.

- Voith LR40 laboratory refiner

A Voith laboratory refiner LR 40 was used to refine pulp sample. The SEL was set at 2.5 J/m to control refining intensity. The Voith LR-40 refining is shown in the figure 27. Voith LR40 has similar refining zone structure as in typical industrial refiner.

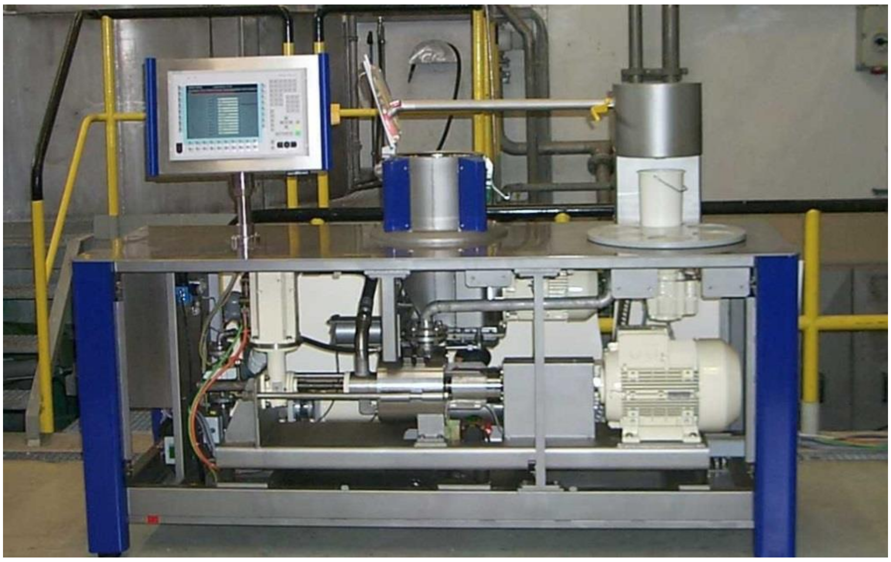


Figure 27. Voith LR-40 Laboratory refiner.

- Masuko Super Masscolloider

Masuko was used to refine pulp in order to create highly external fibrillation on the fiber surfaces. It has two ceramic nonporous grinding surfaces, which are adjustable at any clearance between the upper and lower grinder as shown in figure 28. Masuko is widely used to produce nano/microfibrillated cellulose for lab scale volume.

Figure 28. Masuko Super Masscolloider

Moving Belt Former (MBF)

MBF is equipment used to evaluate dewatering simulated wire section in papermaking. It was developed at Helsinki University of Technology and designed to simulate the pulsating drainage on the wire section of a paper machine. The vacuum level and speed difference between foils and wire are corresponding to production scale Fourdrinier paper machine. The MBF differs from a paper machine that on the MBF because the foil is moving instead of wire. MBF mainly consists of vacuum box connected to a vacuum pump, a moving belt with foils, mesh wire and mixing jar. The vacuum level, the speed of the belt as well can be varied. After drainage a sheet of 19 × 19 cm is obtained. The sheet is then weighed, dried and weighed again and the retention is then calculated. The gained solid content of the wet sheet is obtained as a measure of the drainage. Figure 29 and 30 show the scheme and picture of MBF.

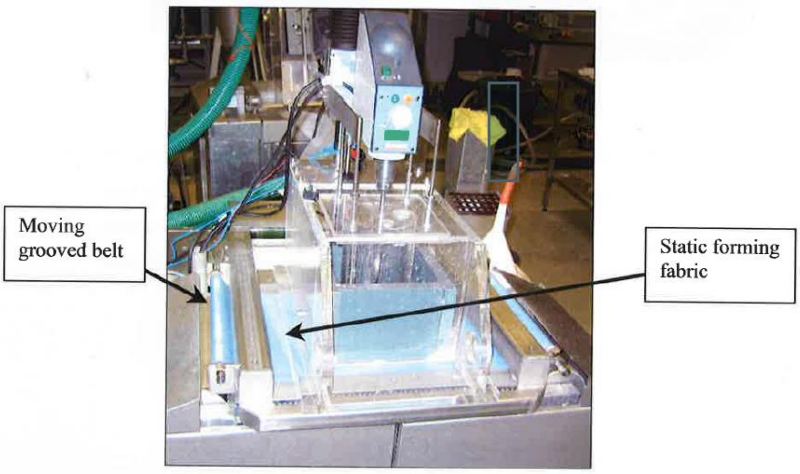


Figure 29. Moving belt former (MBF)

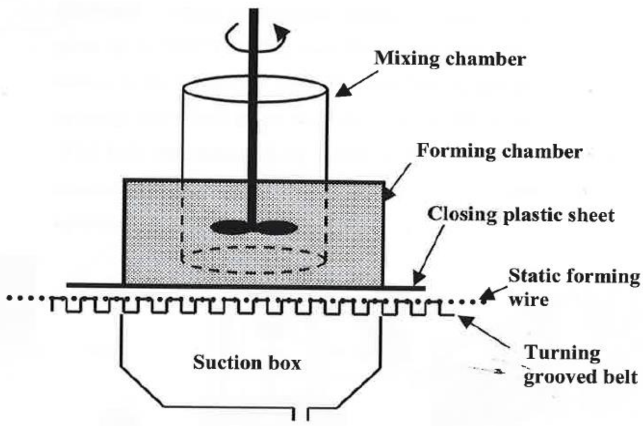


Figure 30. The schematic of Moving belt former (MBF).

Methods

Refining of pulp with Lampén mill and Voith LR40

In Lampén beating, pulp was beaten according to Scan-C23:67 beating of pulp in the Lampén mill, Scandinavian pulp, paper and board testing committee. Pulp was beaten using de-ionized water with 30 oven dry gram per batch, 3% consistency at 3500, 7000, 11000 revolutions

In Voith refining, pulp was refined by Voith LR40 with 3-1, 0-60 3 mm bar width filling with conical design. Pulp was refined using de-ionized water with 60 oven dry grams per sample point, 4% consistency at 0, 75, 150, and 225 kWh/t respectively. Trial protocol was set as follow: dwelling time 10 minutes, pulping time 10 minutes, SEL 2.5 J/m, refining speed 2000 rpm and cutting edge length 0.67 km/s.

Fiber and Pulp Properties evaluation

Evaluation of fibrillation

- Internal fibrillation was evaluated by degree of FSP with method of the solute exclusion according to the studied of Stone, Scallan and et al, 1968.
- External fibrillation was evaluated by Kajaani FS-200 Fiber analyzer. Moreover, the external fibrillation image was taken and speculated from phase contrast microscopy using Leica ICC50 HD with PLAN 10x/0.20 as shown in figure 31.

Figur 31. Leica ICC50 microscope.

Evaluation of Dewatering

Dewatering was evaluated by moving belt former (MBF). The parameter of experiment is as follow: mixing time 30 sec, 2 g/l of pulp consistency, 842 ml of sample volume.

SR and WRV

The Schopper Riegler (SR) was measured according to ISO 5267-1:1999(En) - Determination of drainability - Part 1: Schopper-Riegler method.

Water retention value (WRV) was measured according to SCAN-C 62:00 with Thermo Scientific SL40F/40FR Centrifuge. The Thermo Scientific SL40F/40FR Centrifuge is shown in the figure 33.

Fine Separation

Fine was separated by Britt Dynamic Drainage Jar (BDDJ) according to SCAN-CM 66:05:2005.

Average fiber length (mm), %fine and fiber curl

Fiber analysis was measured with a Kajaani FS-200 Fiber analyzer as shown in figure 32.

Figure 32. Kajaani FS-200 Fiber analyzer at PUU laboratory, Aalto University.

Figure 33. Thermo Scientific SL40F/40FR Centrifuge.

Paper properties testing

All hand sheets were made according to the standard ISO 5269. Deionized water was always used in the experiment.

Strength properties

- Tensile strength was measured according to ISO 1924-2:2008 - Paper and board - Determination of tensile properties - Part 2,3 : Constant rate of elongation method (20, 100 mm/min) by L&W ALWETRON TH1 tensile tester at PUU laboratory, Aalto University.
- Zero span (kN/m) was measured according to ISO 15362:2000 by Pulmac Z-span 100 tester at PUU laboratory, Aalto University.
- Internal bonding strength (J/m2) was measured according to TAPPI T569 pm-00 by Huygen internal bond tester at PUU laboratory, Aalto University.

Optical properties

- All Optical properties, including light scatter, light absorption, ISO Brightness, opacity were measured according to ISO 2471:2008 Paper and board - Determination of opacity (paper backing) - Diffuse reflectance method and ISO 9416:2009 Paper -- Determination of light scattering and absorption coefficients (using Kubelka-Munk theory) by L&W SE070R Elrepho spectrophotometer at PUU laboratory, Aalto University.

Surface properties

- Formation was measured by Ambertec Beta Formation tester at PUU laboratory, Aalto University.
- Roughness was measured according to ISO 8791-2- Determination of roughness/smoothness (air leak methods) - Part 2: Bendtsen method by L&W Bendsten tester at PUU laboratory, Aalto University.

Structural properties

- Thickness and Bulk were measured and calculated according to ISO 534:2011 Paper and board - Determination of thickness, density and specific volume by L&W micrometer at PUU laboratory, Aalto University.

Chapter 4. Results and Discussions

1. Refining VS fiber structural changes

All the measurement results are shown in a table at Appendix 1.

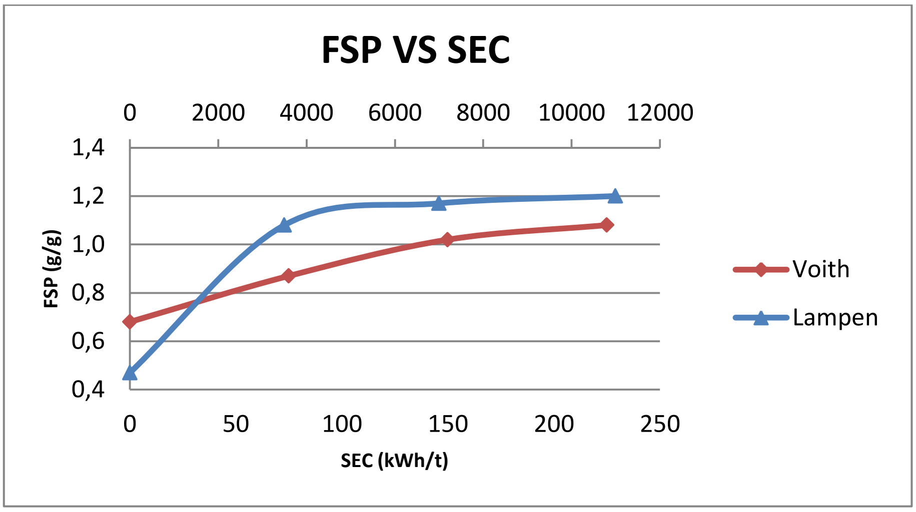


Figure 34. Degree of FSP of softwood pulp beaten by Lampén Mill with 0, 3500, 7000, 11000 revolutions and pulp refined by Voith refiner with 0, 75, 150, 225 kW/t.

In Lampén refining, FSP increased dramatically at low SEC. It rose from 0.47 to 1.08 between 0 and 3500 revolutions. At the higher SEC, FSP only slightly develops. It increased from 1.08 at 3500 revolutions to 1.20 at 11000 revolutions. In Voith refining, FSP increased nearly linearly for the entire SEC range. FSP increased from 0.68 at 0 kWh/t to 1.08 at 225 kWh/t. According to figure 34, FSP of Lampén-beaten pulp is lower than that of Voith-refined pulp before refining because of differences in pulp preparation; however, Lampén-beaten pulp has higher degree of FSP only after light refining. At low SEC, FSP of Lampén-beaten pulp increase more dramatically than that of pulp refined Voith and then increased quite constantly at the higher SEC. Therefore, we could conclude that compressive refining represented by Lampén mill promotes more internal fibrillation than abrasive refining by Voith refiner.

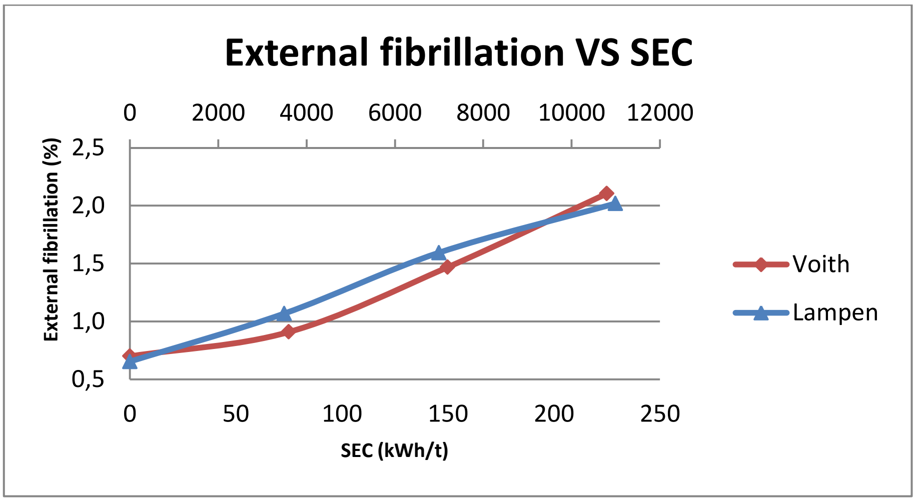


Figure 35. External fibrillation of softwood pulp beaten by Lampén Mill with 0, 3500, 7000, 11000 revolutions and pulp refined by Voith refiner with 0, 75, 150, 225 kWh/t .

External Fibrillation of Lampén-beaten pulp rose almost linearly in the entire refining. It increased from 0.66% to 2.02% between 0 and 11000 revolutions. In Voith-refined pulp, external fibrillation increased slightly at low SEC. It grew from 0.70% to 1.91% from 0 and 75 kWh/t. Then it increases linearly to 2.11% at 225 kWh/t. According to the result from the figure 35, both refining methods could develop large amount of external fibrillations on the fibers. External fibrillations of both pulps refined by Voith refiner and beaten by Lampén Mill grew almost linearly as the SEC increased. The higher the SEC is, the more external fibrillations are generated on fibers.

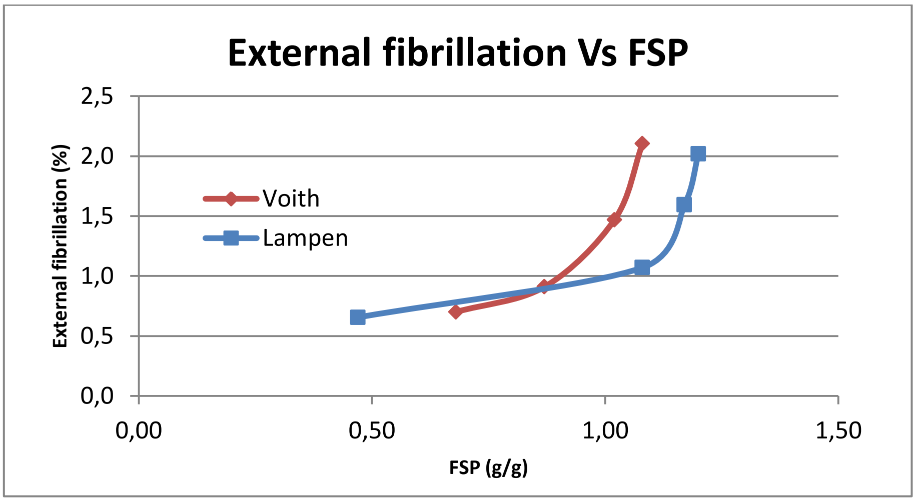


Figure 36a. Results of external fibrillation versus fiber saturation point (FSP) of pulp refined by Voith refiner and beaten by Lampén.

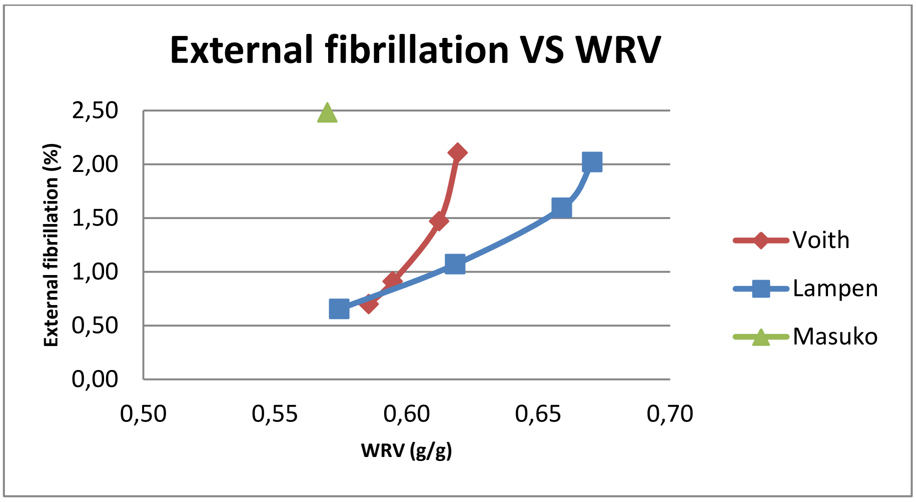


Figure 36b. Results of external fibrillation versus Water retention value (WRV) of Voith-refined pulp, Lampén- beaten pulp and Masuko-refined pulp.

According to the result in figure 36a, internal fibrillations were promoted mostly at the low specific energy whereas external fibrillations were developed mainly at the higher SEC. Both pulps refined by Voith and beaten by Lampén mill had high growth of FSP at low SEC. Then external fibrillation of both pulps increased sharply while FSP only slightly grew at the higher SEC. Lampén refining was able to highly promote external fibrillation which contrasts to some earlier studies (Kang, 2006, Wang, 2007).

In figure 36b, Masuko-refined pulp has extremely high external fibrillation and very low WRV. Even though FSP of Masuko-refined pulp was not measured because of some technical difficulties, according to earlier studies, WRV could be able to roughly evaluate internal fibrillation (Maloney, Laine, Paulapuro, 1999; SCAN-C 62:00, 2000; Kang, 2007). This shows that Masuko-refined pulp has highly developed external fibrillation but little developed internal fibrillation.

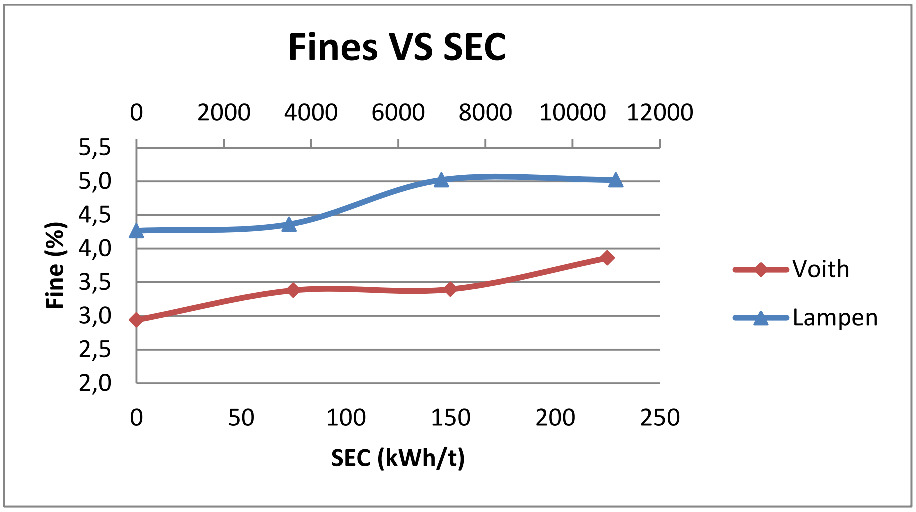


Figure 37. Amount of fines of softwood pulp beaten by Lampén Mill with 0, 3500, 7000, 11000 revolutions and refined by Voith refiner with SEC of 0, 75, 150 and 225 kWh/t.

Fines of Lampén-beaten pulp only slightly increased when refining with high specific energy consumption. It went from 4.27% to 5.02% from 0 to 11000 revolutions. In the case of Voith-refined pulp, %fine increased gradually at the low SEC. It grew from 2.94% to 3.86% from 0 and 225 kWh/t.

The results from the figure 37, illustrate that both compressive and abrasive refining used in the experiment generated only small amount of fine. Amount of fines of both pulp refined by Voith refiner and beaten by Lampén Mill increased less than 1% (1 percent unit) for the entire range of SEC. However, amount of fines of the pulp beaten by Lampén was a little higher than the pulp refined by Voith refiner at zero and any SEC. The reason for this is probably the differences of disintegration devices. Nevertheless, please note that Kajaani fiber analyzer apparatus has limited potential to measure fines. It can only detect at the certain range of the length of fine.

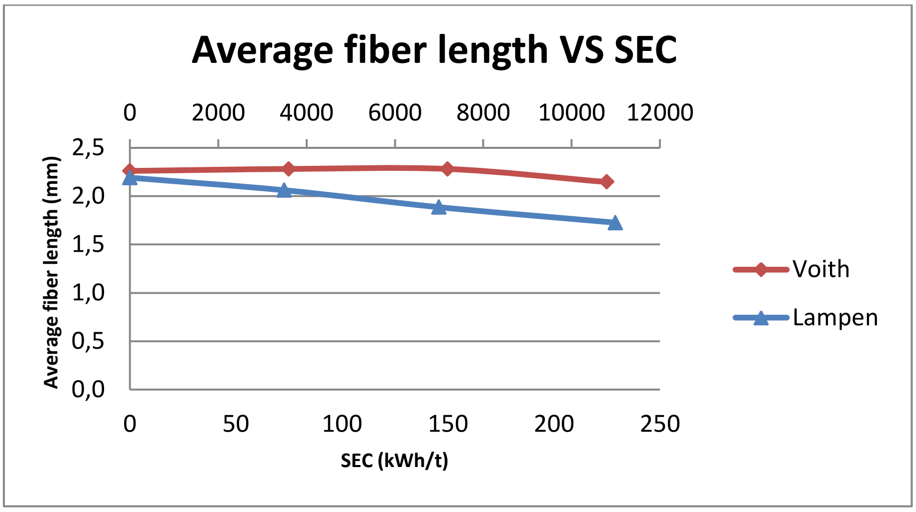


Figure 38. Average fiber length of softwood pulp refined by Lampén with 0, 3500, 7000, 11000 revolutions and pulp refined by Voith refiner with 0, 75, 150, and 225 kWh/t.

The average fiber length significantly decreased when pulp was beaten by Lampén mill with high revolutions. It decreased about 21% from 2.19 mm to 1.73 mm from 0 to 11000 revolutions. The average fiber length of Voith-refined pulp remained almost constant at 2.28 mm from 0 until 150kWh/t and slightly decreased to 2.15 mm at 225 kWh/t.

According to the figure 38, fibers of the pulp beaten by Lampén were shortened more severely than those of the Voith-refined pulp. In Voith refining, the intensity or specific edge load (SEL) used in the experiment was moderate (2.5 J/m) for pulp which has long fiber. However, the intensity was high enough to develop tensile strength index well (shown in Appendix 1.). On the other hand, fiber from Lampén beaten pulp were shorten because of tensile force built up in the beating process due to “hammering effect” between Lampén ball and fiber flocs. Hammering effect in this study refers to the impulsive force hitting between two different objects. Impacts of Hammering effect were heard as loud noises during Lampén refining.

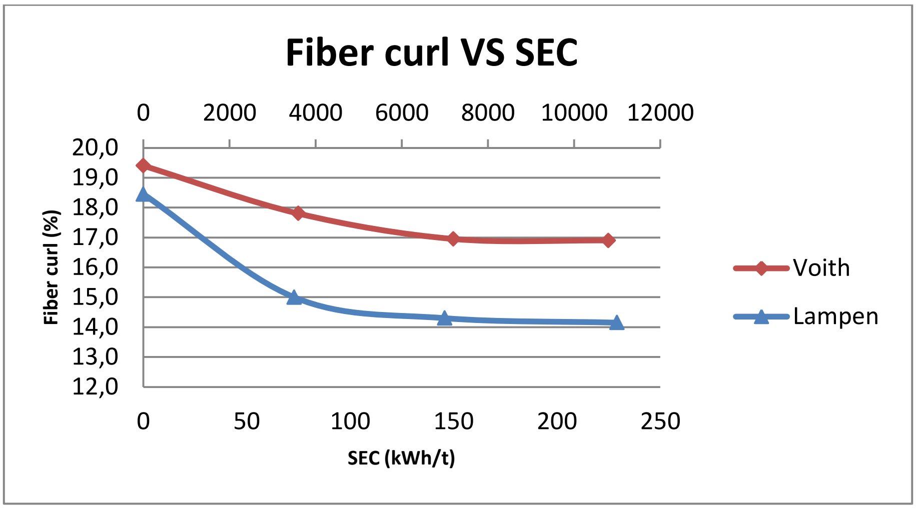


Figure 39a. Degree of Fiber curl of softwood pulp beaten by Lampén Mill with 0, 3500, 7000, 11000 revolutions and pulp refined by Voith refiner with 0, 75, 150 and 225 kWh/t.

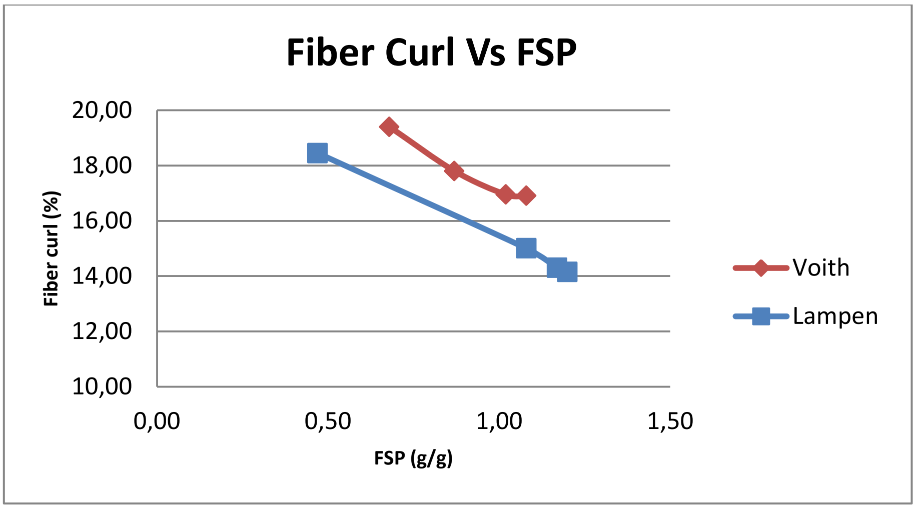


Figure 39b. Results of fiber curl versus fiber saturation point (FSP) of pulp refined by Voith refiner and beaten by Lampén.

Fiber curl of Lampén-beaten pulp decreased dramatically at low SEC meaning that the fibers were largely straightened. It declined from 18.45% to 15.00% between 0 and 3500 revolutions. At the higher SEC, fibers were slightly straightened. The curl index decreases to 14.15% at 11000 revolutions. The result from Voith-refined pulp presented similar trend. Fiber curl declined gradually from 19.40% at 0 kWh/t to 16.95% at 150 kWh/t. Then fiber curl slightly decreased to 16.90% at 225 kWh/t. According to the figure 39a, both fibers of the pulp refined by Voith refiner and beaten by Lampén were straightened much more effectively in the beginning of refining than the later refining. However, fibers were likely to be straighter after beaten by Lampén mill than fiber refined by Voith refiner. Fiber straightening in Lampén refining might be caused by tensile force built up during refining based on the idea of Hammering effect.

Figure 39b shows that fiber curl and FSP have a linear correlation indicating that fiber curl and internal fibrillation developed simultaneously.

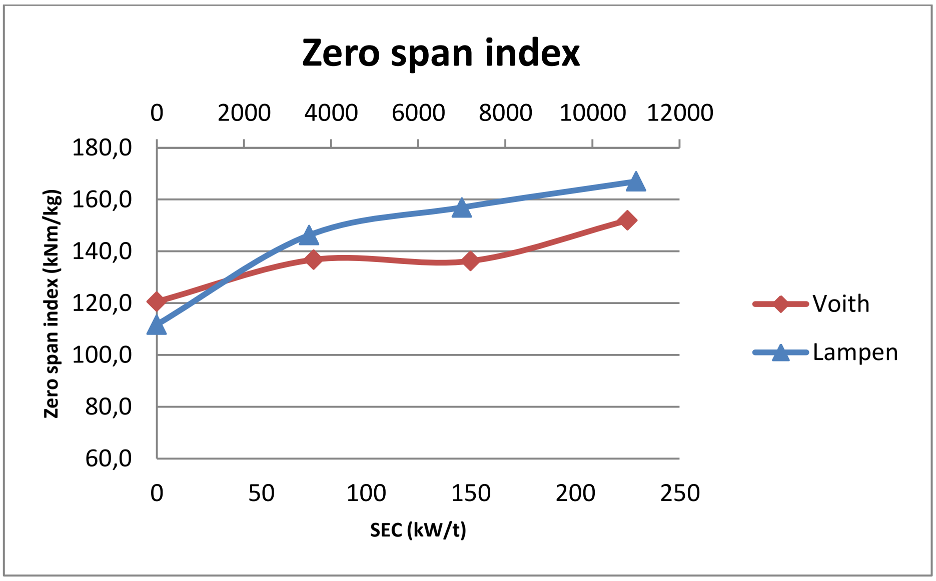


Figure 40. Effect of Lampén beating and Voith refining on zero span breaking strength of softwood pulp.

Zero span from Lampén-beaten sample increased steadily approximately 49.5%. It rose from 111.7 to 167.0 kNm/kg between 0 and 11000 revolutions. In Voith refining, zero span increased approximately 26.0%. It grew from 120.6 kNm/kg to 151.9 kNm/kg between 0 and 225 kWh/t. According to the figure 40 from the experiment, zero spans were moderately increased by both refining method. Even though zero span is widely used to determine average fiber strength, it is difficult to assess whether the development result from fiber strength or inter bonding between fiber. However, according to Wathén's study, the reason to explain the increasing of zero span could be because of the favorable organization of microfibrils during refining (Wathén, 2006). Please note that the grip separation used in the zero span measurement is theoretically 0 mm.

2. Fiber structural changes VS Paper properties

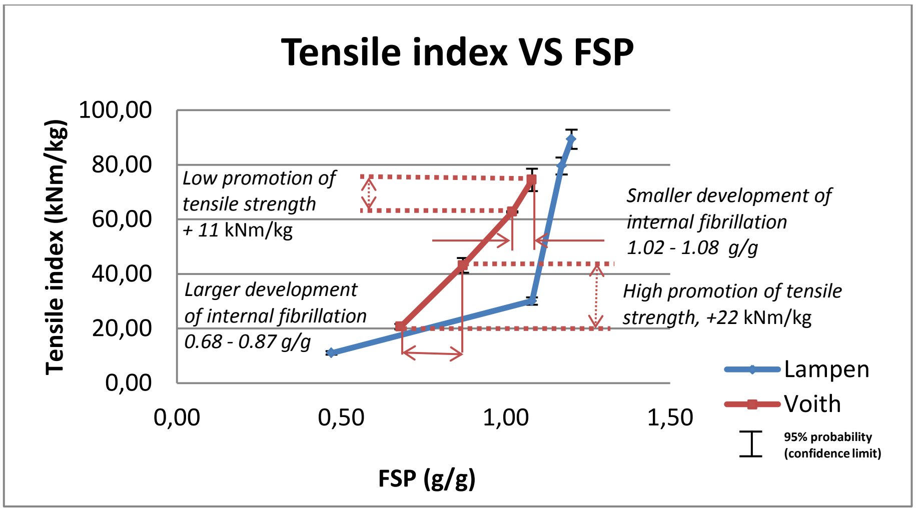


Figure 41a. Effect of FSP on tensile index of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

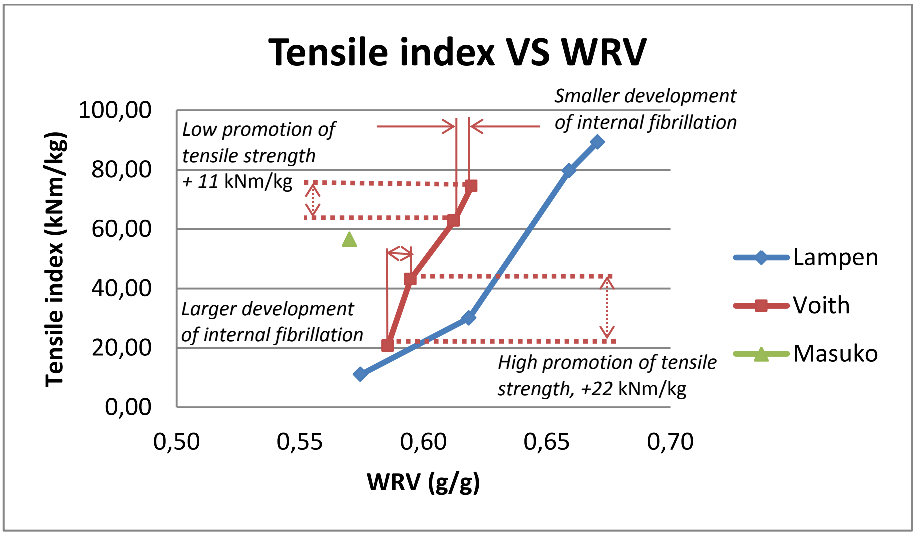


Figure 41b. Effect of WRV on tensile index of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

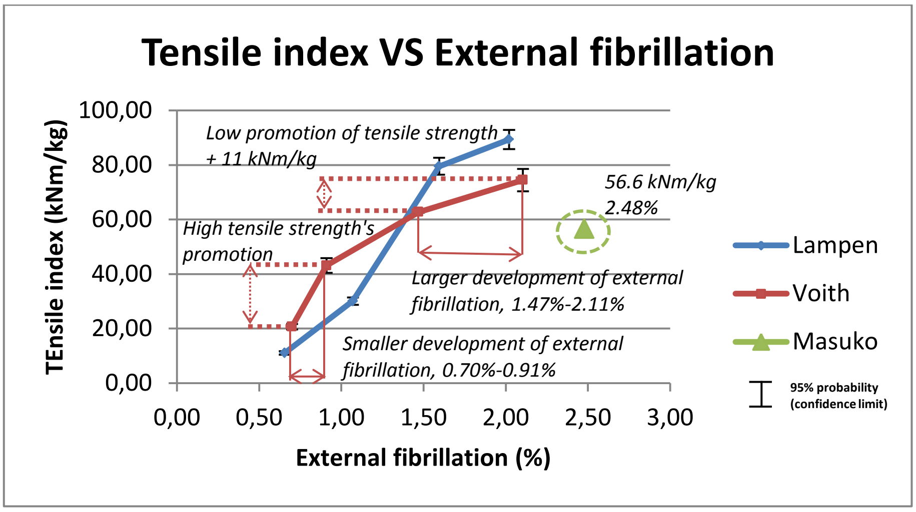


Figure 42. Effect of external fibrillation on tensile index of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

Figure 41a, 41b and 42 illustrate the effect of internal and external fibrillations on tensile strength. It was obvious that both internal and external fibrillations significantly promote tensile strength. In both Lampén and Voith refining methods, tensile strength was promoted more substantially at low SEC where internal fibrillation was highly developed whereas the tensile strength was less promoted at the higher SEC where external fibrillation was more developed. According to figure 41a and 42, in Voith refining at range of 0.68 - 0.87 g/g and 0.70% - 0.91%, which represented the range of internal fibrillation's development, tensile strength index increased approximately 22 kNm/kg (from 21 to 43 kNm/kg) while in the higher range of 1.02 - 1.08 g/g and 1.47% - 2.11% which represented the higher development of external fibrillation, tensile strength index increased only 11 kNm/kg (from 74 to 63 kNm/kg). The effect of internal and external fibrillation from Lampén refining on tensile strength illustrated the same results when analyzing with the same method. Therefore, it was evident that internal fibrillation affect tensile strength more crucial than external fibrillation.

The conclusion was confirmed by the result of Masuko-refined pulp in figure 41b and 42. Tensile strength of Masuko-refined sample was just moderately improved; even though, external fibrillations were highly developed.

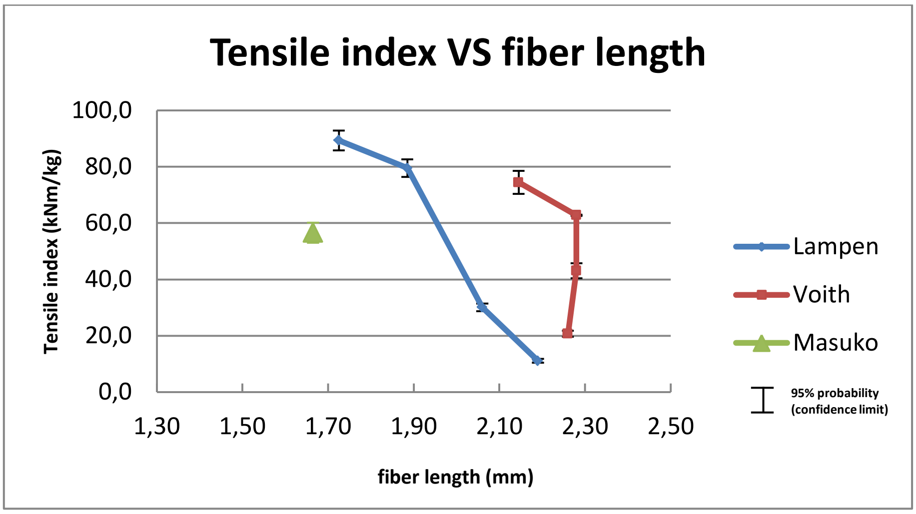


Figure 43. Effect of fiber length on tensile index of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

Figure 43 shows that fiber length did not correlate with tensile strength. At the same tensile strength index, each sample had different fiber length. For example, at 56 kNm/kg, fiber length of Masuko-beaten sample was 1.67 mm whereas fiber length of Lampén and voith-refined sample was aprroximately 1.95 and 2.28 mm repectively.

Fiber curl and internal fibrillation were developed simultaneously throughout the refining in both refining methods (shown in figure 39b) so that it was difficult to assess their relative importance in property development.

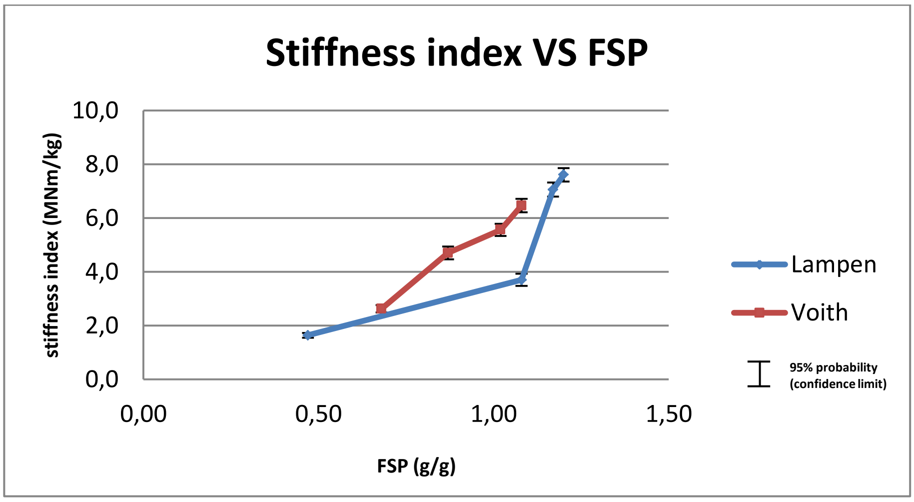


Figure 44. Effect of FSP on tensile stiffness index of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

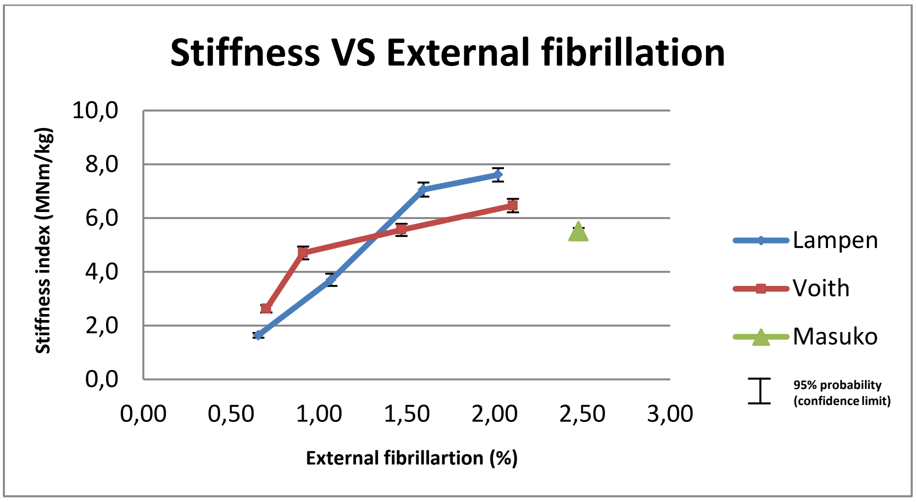


Figure 45. Effect of external fibrillation on tensile stiffness index of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

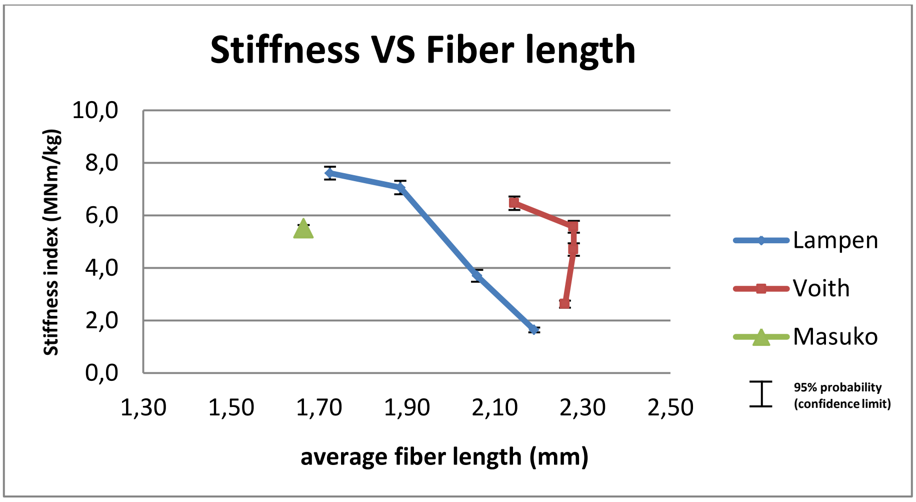


Figure 46. Effect of fiber length on tensile stiffness index of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

Figure 44, 45, 46 show the effect of structural changes of fiber on tensile stiffness (i.e. specific elastic modulus). The result of tensile stiffness presented the similar trend as tensile strength index. It could be that structural changes on fiber affected tensile stiffness in the same way as tensile strength.

Both internal and external fibrillations promoted tensile stiffness but internal fibrillation had higher effect on the stiffness resulting in more rigid paper sheet. In addition, fiber length had ambiguous correlation with the improvement of stiffness.

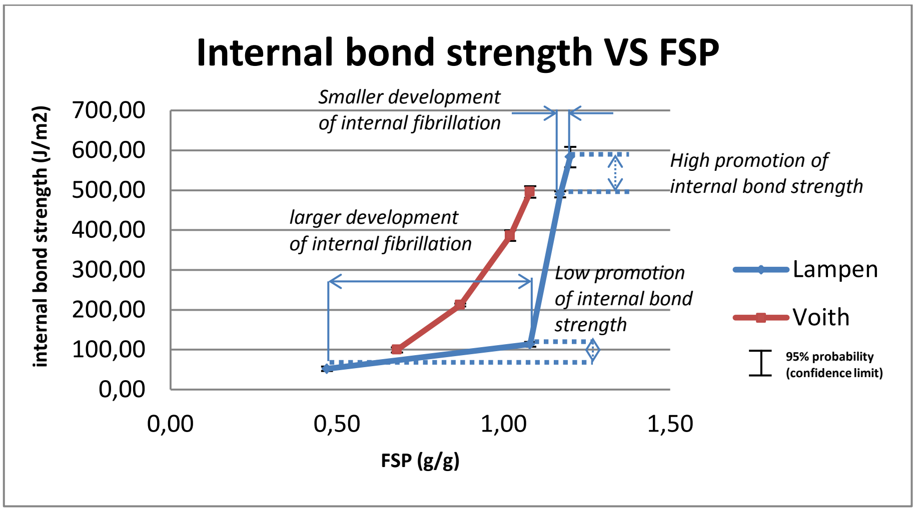


Figure 47. Effect of internal bonding strength on internal bonding strength of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

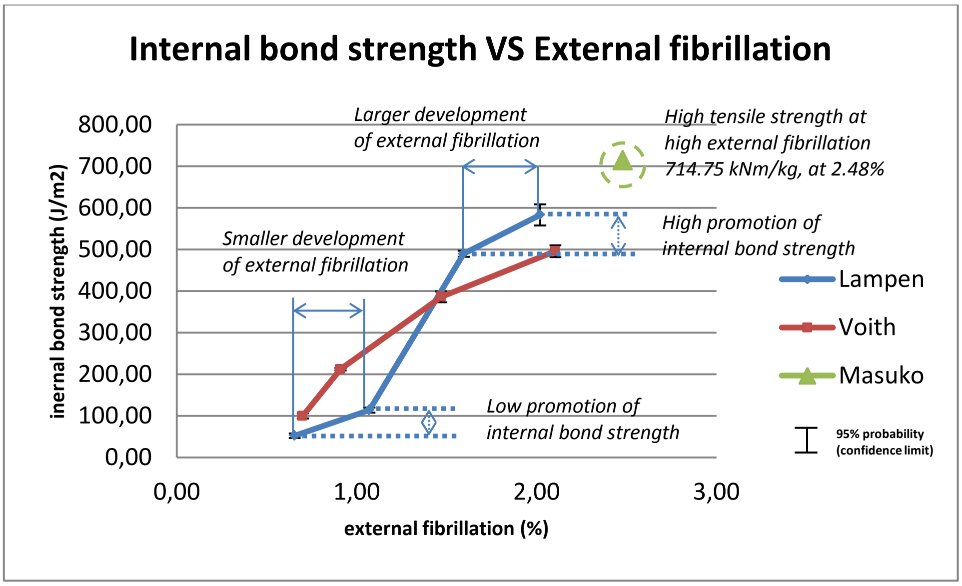


Figure 48. Effect of external fibrillation on internal bonding strength of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

Figure 47 and 48 illustrate the effect of internal and external fibrillation on internal bonding strength. It was evident that both internal and external fibrillations developed internal bonding strength; however, the effect of external fibrillation was higher than that of internal fibrillation. Similar explanation method as tensile strength could be described for internal bond strength.

According to figure 47 and 48, in Lampén beating at range of 0.47 - 1.08 g/g and 0.66% - 1.07%, which represented the range of internal fibrillation's development, internal bond strength increased only 62 kNm/kg (from 52 to 114 kNm/kg) while in the higher range of 1.17 - 1.20 g/g and 1.60% - 2.02% which represented the development of external fibrillation, internal bond strength increased 93 kNm/kg (from 490 to 583 kNm/kg). Therefore, it was evident that external fibrillation affected internal bond strength more crucial than internal fibrillation. This was supported by the result of Masuko-refined pulp. Internal bond strength of Masuko-refined sample whose external fibrillations were highly developed was extremely developed.

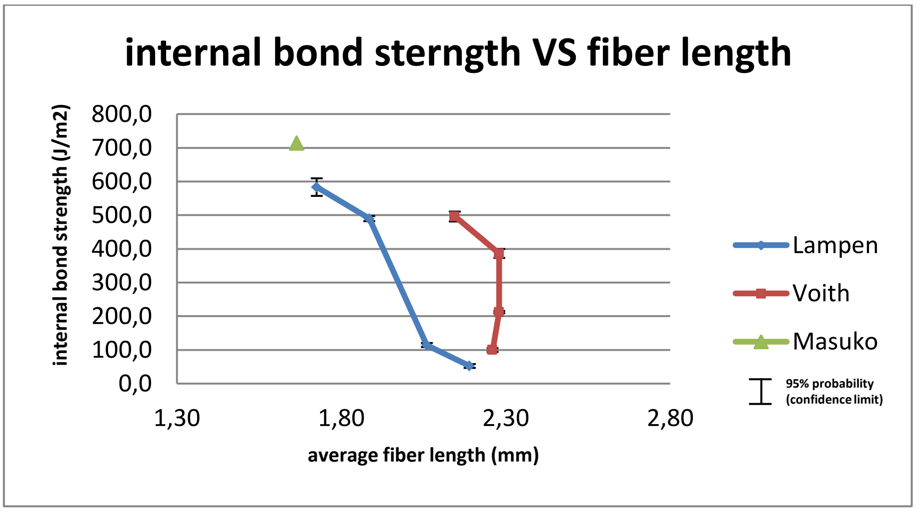


Figure 49. Effect of fiber length on internal bonding strength of sample sheets from Lampén-beaten and Voith-refined.

Figure 49 shows that fiber length affected internal bonding strenght slightly. The results from the Masuko and Lampén sample tended to correlate well with internal bonding strenght that the shorter the fibers was, the greater the significance of bonding became. Consequently, the fiber shortening benefited z-direction strength properties.

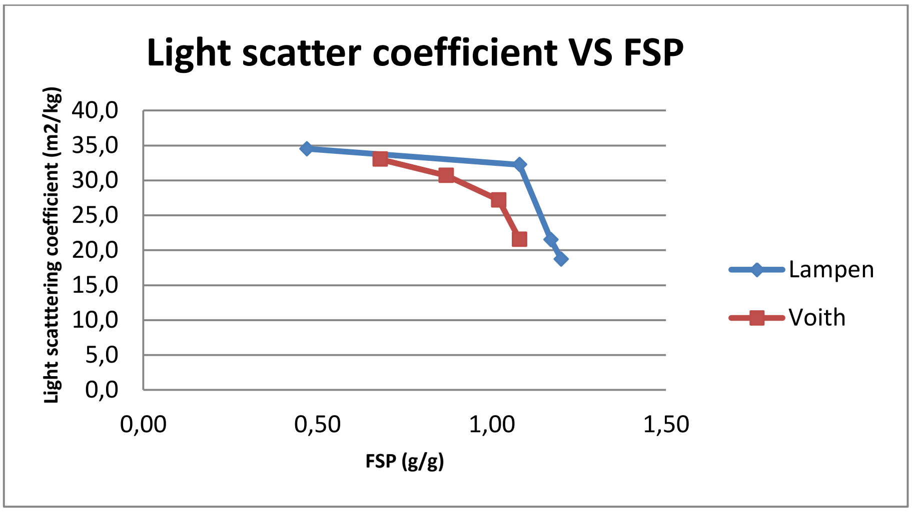


Figure 50. Effect of internal bonding strength on light scattering coefficient of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

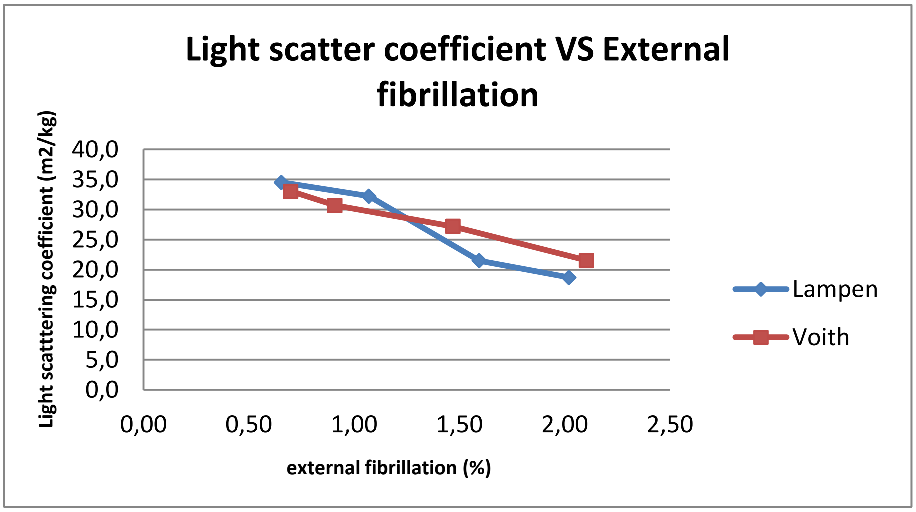


Figure 51. Effect of external fibrillation on light scattering coefficient of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

Figure 50 and 51 illustrate the effect of internal as well as external fibrillation and light scattering. It was evident that both internal and external fibrillation decreased light scattering coefficient; however, the effect of external fibrillation was larger than that of internal fibrillation. Light scattering coefficient declined more greatly at the higher SEC where external fibrillation was highly promoted.

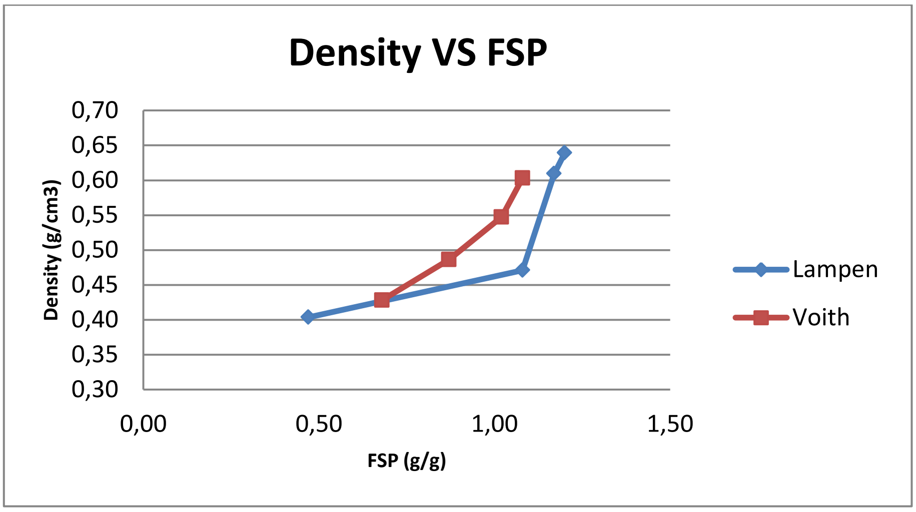


Figure 52. Effect of internal fibrillation on density coefficient of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

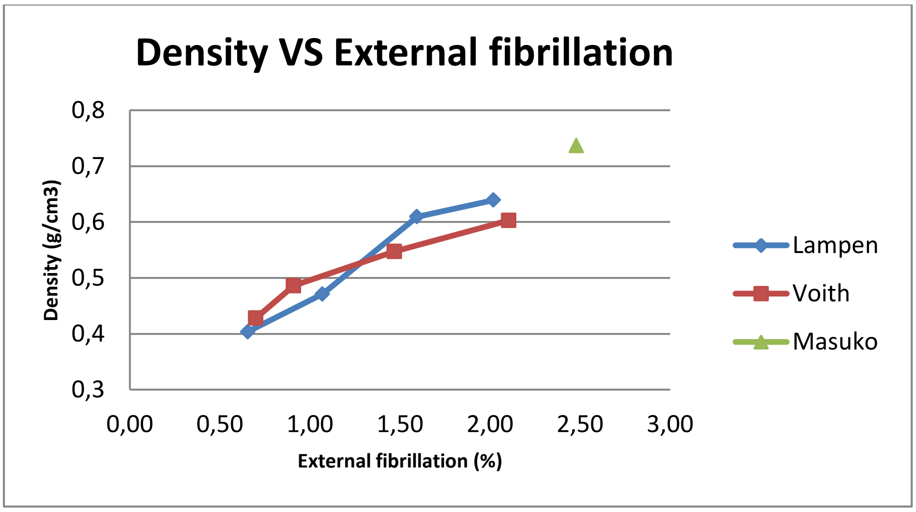


Figure 53. Effect of external bonding strength on density of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

Accoding to the result in figure 52 and 53, it was obvious that internal and external fibrillation strongly promoted density. The result correlates well with the early study that the greater external fibrillation is, the higher the density of the sheet sample become. Increasing in relative bonded area caused by external fibrillation lowered porosity of sheet resulting in higher density. Moreover, due to the fact that internal bonding increses fiber swelling, the fiber tend to shrink more in the process of drying.

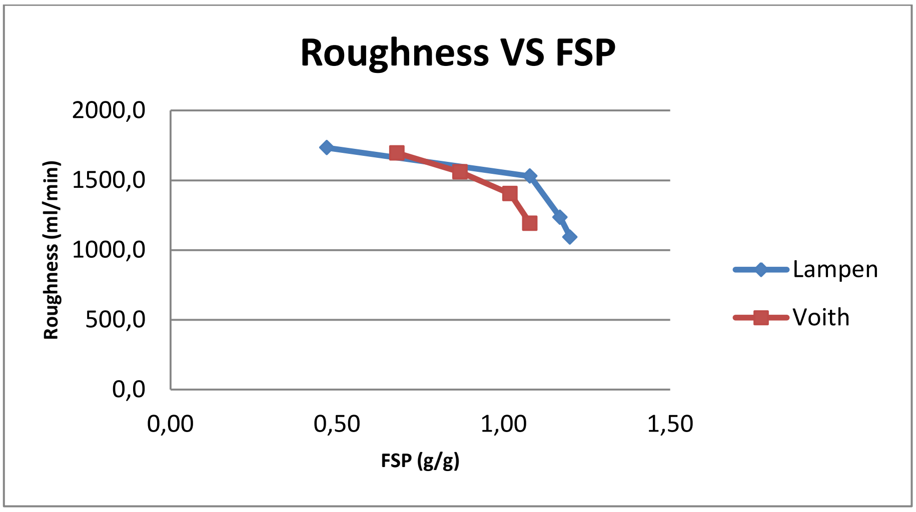


Figure 54. Effect of internal fibrillation on roughness of sample sheets from Lampén-beaten and Voith-refined pulps.

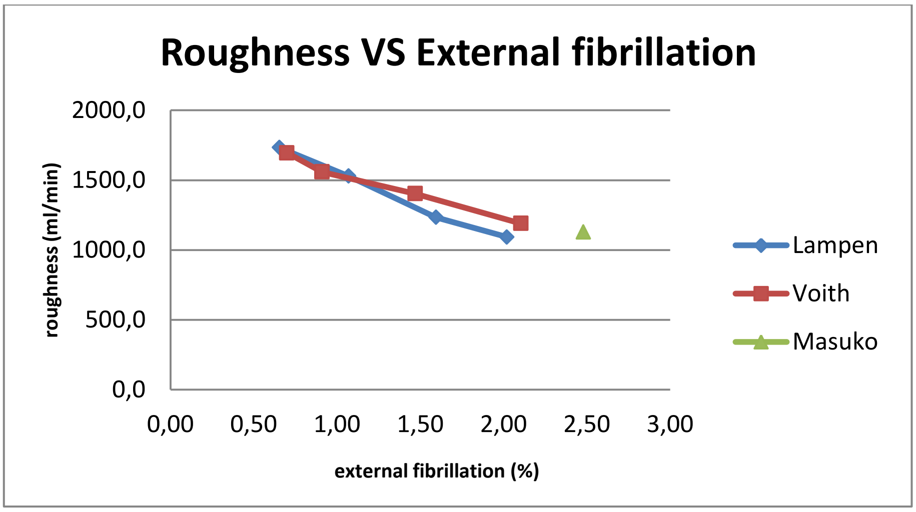


Figure 55. Effect of external fibrillation on roughness of sample sheets from Lampén-beaten, Voith-refined and Masuko-refined pulps.

Accoding to the result in figure 54 and 55, it was obvious that external fibrillation greatly decreased roughness. The result illustrated that the higher external fibrillation was, the lower the roughness of the sheet sample became. Increasing in bonding ability lowered porosity of sheet resulting in smoother suface. Moreover, according to the result, internal fibrillation also increased smoothness of fiber surface.

3. Refining type VS Paper properties

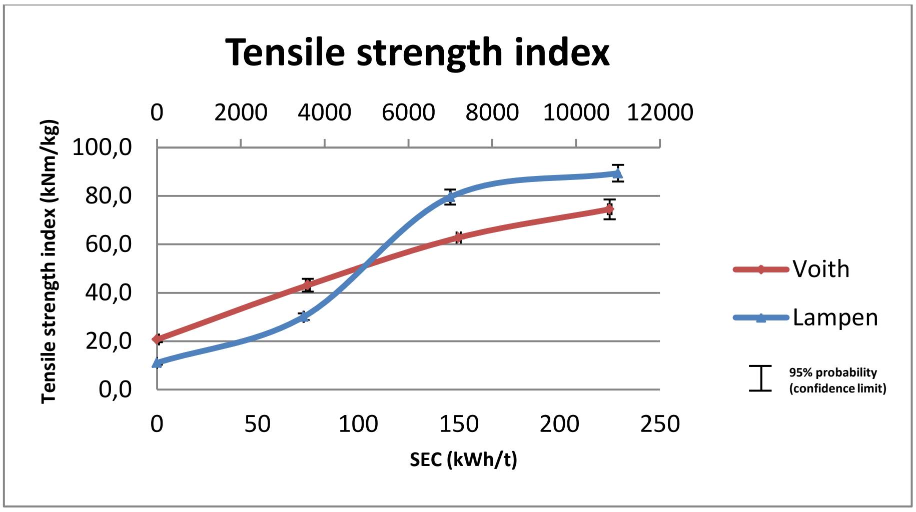


Figure 56. Tensile index of softwood pulp beaten by Lampén Mill with 0, 3500, 7000, 11000 revolutions and pulp refined by Voith refiner with 0, 75, 150, 225 kWh/t.

According to figure 56, tensile index of Lampén-beaten sample at 3500 revolutions nearly tripled the tensile index of unrefined pulp. Then it increased even more sharply between 3500 and 7500 revolutions. It rose from 30.07 kNm/kg to 79.56 kNm/kg. At the higher SEC, the tensile index only slightly developed. It increased to 89.36 kNm/kg at 11000 revolutions. From Voith refining, tensile index increased nearly linearly from 20.68 kNm/kg at 0 kWh/t to 74.47 kNm/kg at 225 kWh/t.

The result illustrated that tensile strength was able to largely improve by both refining methods. Even though tensile index of sheet samples from both refining increased largely according to the result, the tensile index from Lampén refining grew more steeply in the middle of the refining process when internal fibrillation was greater promoted.

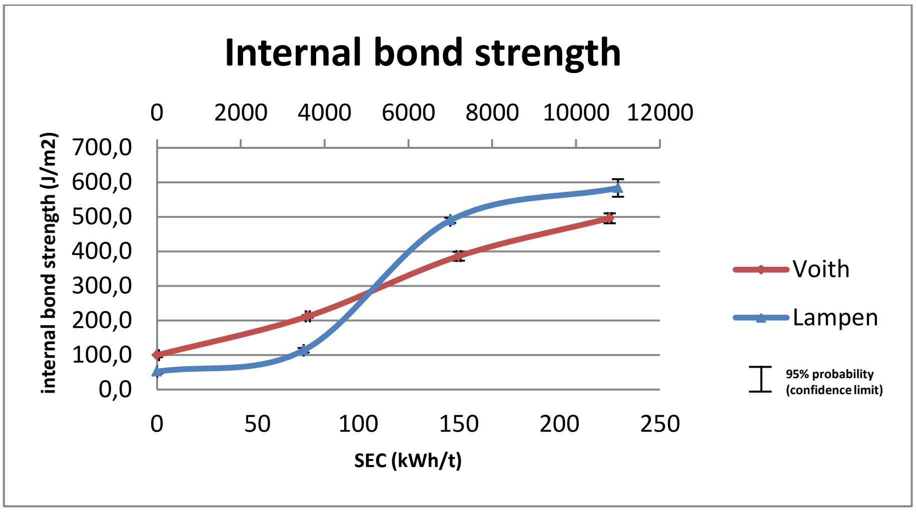


Figure 57. Effect of Lampén beating (0, 3500, 7000, 11000 revolutions) and Voith refining (0, 75, 150, 225 kWh/t) on internal bond strength of softwood pulp.

According to figure 57, internal bond strength from Lampén-beaten sample slightly increased in the low SEC range. However, it increased substantially in the middle of refining between 3500 and 7500 revolutions. It rose from 114 kNm/kg to 490 J/m^2 and ended up at 583 J/m^2 at 11000 revolutions. In Voith refining, it increased nearly linearly from 100 J/m^2 at 0 kWh/t to 496 J/m^2 at 225 kWh/t. The results illustrated that internal bond strength were largely developed by both refining methods. However, the internal bond strength of sample beaten by Lampén increased the most dramatically in the middle of the refining process after internal fibrillation was almost fully developed.

The result correlates well with early study (Wang, 2006; Kang, 2007). Both refining methods promoted fiber shortening and external fibrillation which reduced porosity and improved z-direction bonding resulting in higher internal bonding strength.

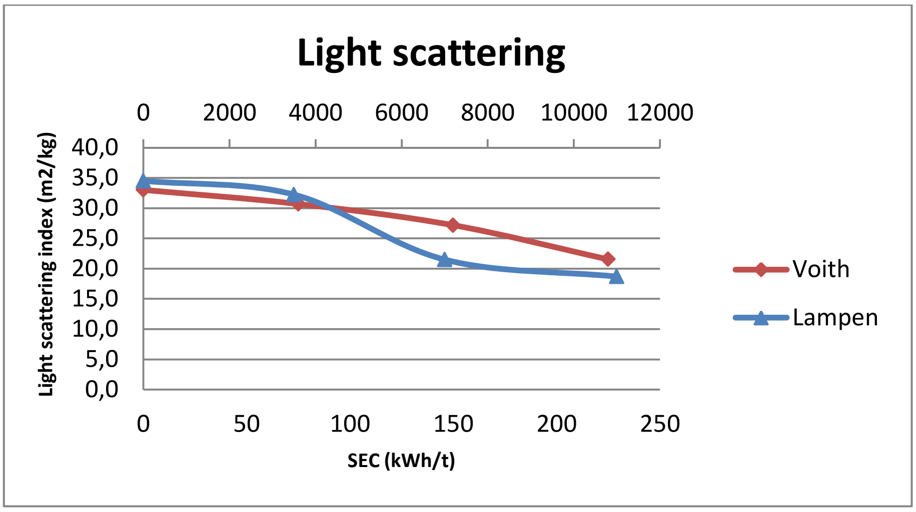


Figure 58. Effect of Lampén beating (0, 3500, 7000, 11000 revolutions) and Voith refining (0, 75, 150, 225 kWh/t) on light scattering coefficient of softwood pulp.

According to figure 58, light scattering of Voith-refined sample decreased almost linearly while that of Lampén-beaten sample dropped largely during the middle of SEC. In Lampén beating, light scattering coefficient continuously decreased when refining with higher revolution starting from 34.5 m^2/J to 18.7 m^2/J between 0 and 11000 revolutions. It decreased significantly in the middle of the refining between 3500 and 7000 revolutions. On the other hand, the light scattering coefficient of Voith-refined sample steadily declined from 33.0 m^2/J to 21.5 m^2/J between 0 and 225 kWh/t. The results agree well with early study that refining decreases light scattering coefficient due to the denser structural network.

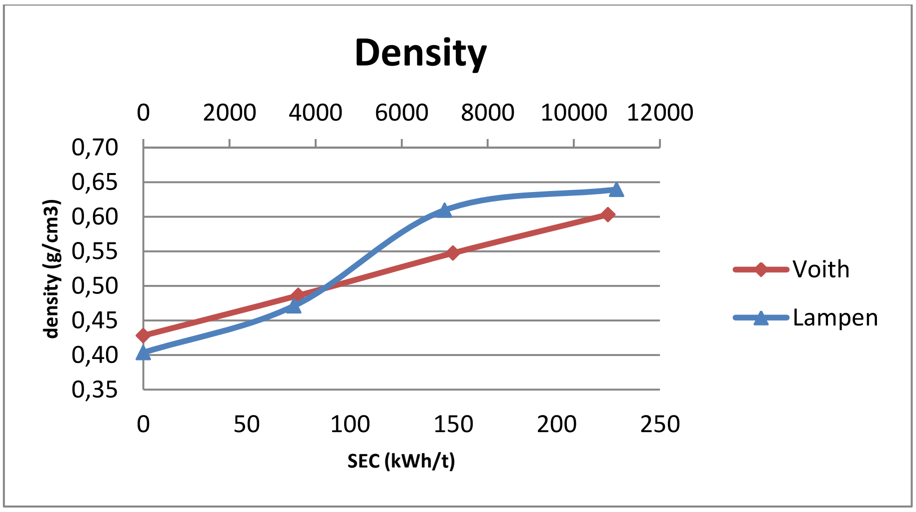


Figure 59. Effect of Lampén beating and Voith refining on density of softwood pulp.

According to figure 59, the result illustrated that the density was significantly developed by both refining methods. The density of sheet sample refined by Voith refiner was developed almost linearly while the internal bonding strength of sample beaten by Lampén increased more dramatically in the middle of the refining process. The density of Lampén-beaten sample increased more substantially in the middle of SEC to 0.61 at 7500 revolutions. After that the density slightly rose to 0.64 g/cm^2 at 11000 revolutions. In Lampén beating, the density increased from 0.43 g/cm^2 at 0 kWh/t to 0.60 g/cm^2 between 0 and 225 kWh/t. The result corresponds to the early study that refining usually increases density. Fiber shortening and external fibrillation from the refining reduce porosity and promote inter-fiber bonding. Moreover, internal fibrillation improves fiber flexibility which finally increases fiber swelling and shrinkage. As a result, these reactions make fiber network more compact.

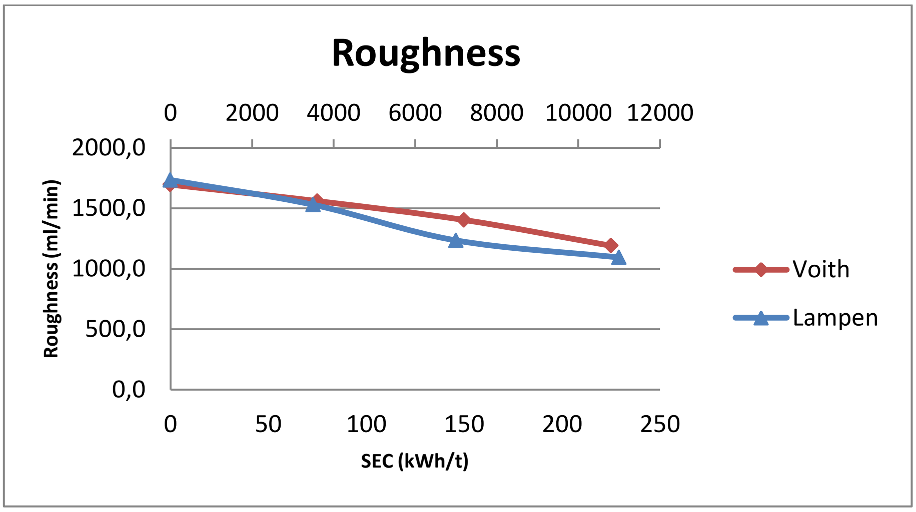


Figure 60. Effect of Lampén beating (0, 3500, 7000, 11000 revolutions) and Voith refining (0, 75, 150, 225 kWh/t) on roughness of softwood pulp.

According to figure 60, the result illustrated that the roughness was largely decreased by both refining methods with nearly linear trend as the SEC increases. The result corresponds to the early study that refining improves surface smoothness of paper. Fiber shortening and external fibrillation from the refining reduce porosity and promote inter-fiber bonding.

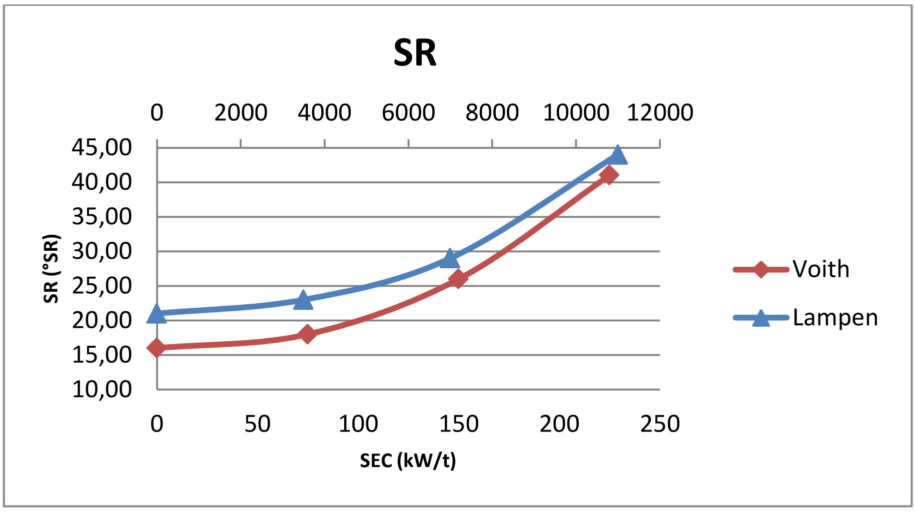


Figure 61. Effect of Lampén and Voith refining on SR.

According to figure 61, the result illustrated that the SR was largely increased by both refining methods with similar trend as the SEC increases. The result corresponds to the early study that refining develops SR and decrease dewatering efficiency.

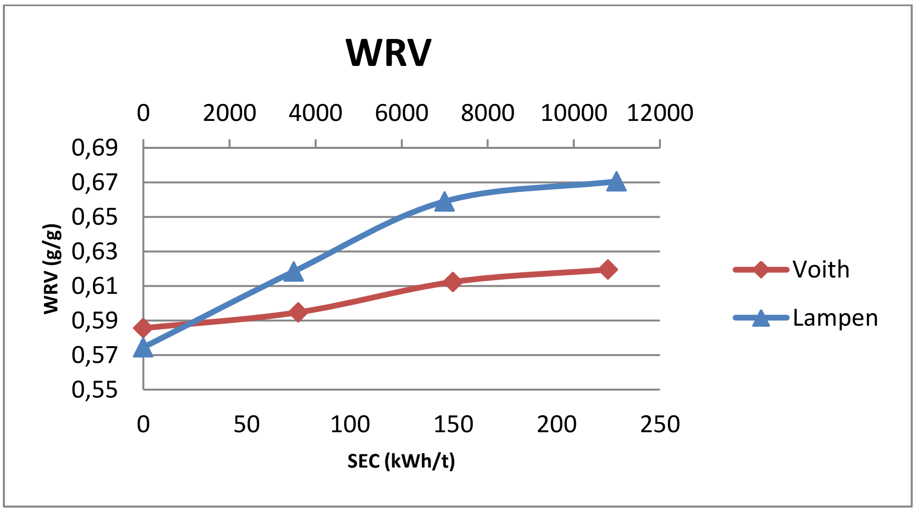


Figure 62. Effect of Lampén and Voith refining on WRV.

According to figure 62, the result illustrated that refining increase WRV in both refining methods; however, Lampén increases WRV more than Voith refining. The result corresponds to the early study that refining promote bonding competency between fiber structure and free water as a result of the growth of internal fibrillation and external fibrillation (Kajanto and Niskanen, 2000).

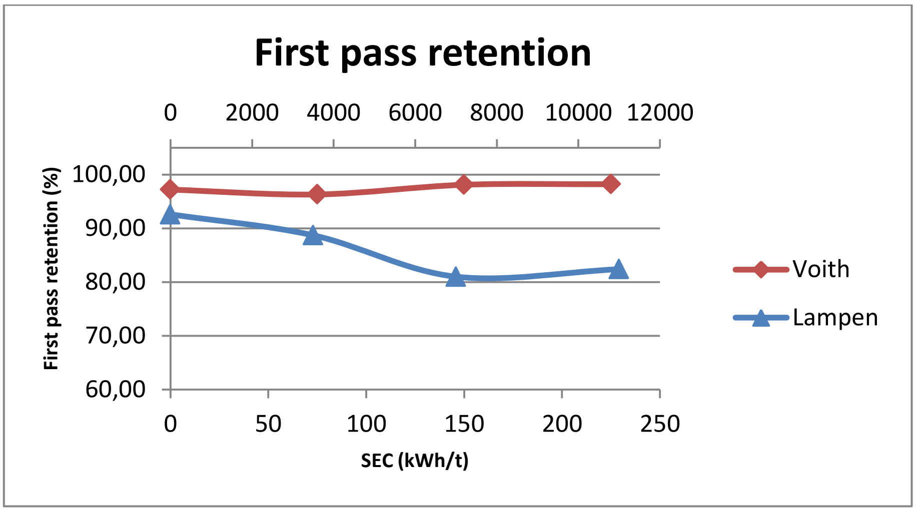


Figure 63. Effect of Lampén beating and Voith refining on first pass retention of softwood pulp. The data was conducted using moving belt former (MBF).

According to figure 63, pulp refined by Voith refiner had constant first pass retention while pulp refined by Lampén has lower first pass retention at high SEC. On the one hand, first pass retention of Voith sample nearly remained constant between 96.30 and 98.10%. It's partially because of the higher fines amount. Nevertheless, the reason why first pass retention of the Lampén sample slightly decreased is unknown.

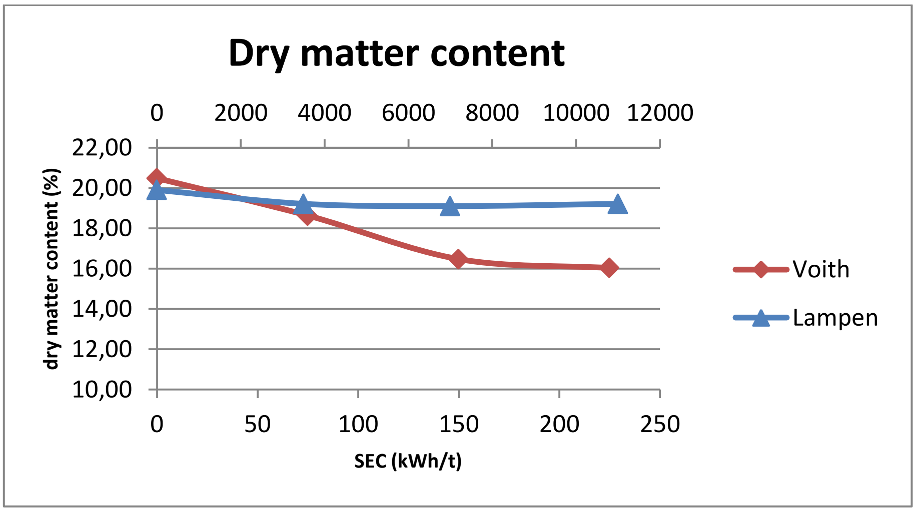


Figure 64. Effect of Lampén beating and Voith refining on dewatering efficiency of softwood pulp. The data was conducted using moving belt former (MBF).

The figure 64 illustrated that dry matter content of pulp refined by Voith refiner dropped at high SEC meaning that Voith refining decreased dewatering efficiency of pulp. The dry matter content of pulp refined by Voith refiner continuously declined from 20.48 to 16.47 at 150 kWh/t and then slightly dropped to 16.03% at 225 kWh/t. The reason of this could be partially explained by the higher fines amount. On the other hand, the dry matter content of pulp beaten by Lampén almost remained constant the entire SEC meaning that Lampén refining had no effect on dewatering efficiency. From figure 61, 62 and 64, MBF ranked Lampén and Voith refining in the opposite order compared to SR and WRV. Since MBF provides rather realistic water removal condition, this demonstrates the unreliability of SR and WRV as indicators for wire section dewatering.

4. Summary of the results

Effect of refining type on paper properties

Table 2. Summary of Effect of refining type on paper properties

	Voith	**Lampén**	**Masuko**
Tensile strength	Highly increase	Extremely increase	Moderately increase
Stiffness	Highly increase	Extremely increase	Moderately increase
internal bond strength	Highly increase	Highly increase	Extremely increase
Light scattering	Highly decrease	Highly decrease	Moderately decrease
Density	Highly denser	Highly denser	Extremely denser
Smoothness	Highly smoother	Highly smoother	Highly smoother

It is apparent that both Lampén mill (compressive refining) and Voith refiner (combination between abrasive and compressive refining) strongly promotes paper strength, including tensile strength, stiffness and internal bonding strength; however, the sample beaten by Lampén shows slightly higher development on tensile strength and stiffness at high SEC.

The result of light scattering coefficient corresponds to the previous study that refining decreases light scattering coefficient. Both refining methods have high level of external fibrillation resulting in the growth of bindings between fibers so that the light penetrating sheet does not scatter.

In structural property, both methods of refining greatly increase density because of the expansion of RBA resulting from the growth of external fibrillation. However, Lampén-beaten sample has slightly denser structure due to the higher degree of fiber flexibility resulting from the development of internal fibrillation.

All refining methods greatly improve smoothness of sheet surface due to the high growth of external fibrillation. Lampén-beaten sample has slightly smoother surface because of superior fiber shrinkage ability.

Effect of refining type on structural changes in fiber

Table 3. Summary of effect of refining type on structural changes in fiber

	Voith	Lampén	Masuko
internal fibrillation	moderately develop (based on FSP result)	highly develop (based on FSP result)	slightly develop (based on WRV result)
external fibrillation	highly develop	highly develop	Extremely develop
Fiber curl	less straight	more straight	least straight
fiber length	longer	shorter	shortest
% fine	low	low	high
Zero span	highly develop	highly develop	slightly develop

All refining methods generate various changes in fiber structure, including internal fibrillation, external fibrillation, fines formation and fiber straightening and fiber cutting. Compressive refining generates higher degree of internal fibrillation and fiber straightening whereas abrasive refining creates external fibrillation more effectively. Average fiber length of Lampén mill-beaten pulp is shorter than those of Voith-refined pulp showing that the refining intensity of Lampén mill used in the experiment is higher than Voith refiner. Both Lampén and Voith refining significantly improve zero span breaking strength which implies the increase of average individual fiber strength and favorable fiber orientation. Internal fibrillation was mostly developed at low SEC area followed by external fibrillation at the higher SEC area in both Lampén and Voith refining. Moreover, internal fibrillation and fiber curl show the similar developing trend in both refining.

Both Lampén beating and Voith refining created only little fines meaning that specific edge load (refining intensity) used in the experiment was relatively low. However, Lampén beating shorten fiber more than Voith refining. Fiber shortening and fiber straightening in Lampén refining can be explained by tensile force built up in the beating process due to the effect that the heavy beating ball impulsively hit fiber flocs.

Masuko refining is determined to highly promote external fibrillation and fiber cutting while it only slightly develops internal fibrillation, zero span and fiber straightening. Lampén mill is also found to be able to develop high degree of external fibrillation when refining with high SEC which contrast to the previous study that Lampén beating does not externally fibrillate fiber (Kang, 2006, Wang, 2007).

Effect of structural changes in fiber on paper properties

Table 4. Summary of effect of structural changes in fiber on paper properties

	Strength properties			Optical property	Surface properties	Structural property
	Tensile strength	**Tensile stiffness**	**Internal bonding**	**Light scattering**	**Roughness**	**Density**
Internal fibrillation	extremely high effect	extremely high effect	Low effect	Low effect	Low effect	Low effect
	increase	increase	increase	decrease	smoother	more compacted
External fibrillation	high effect	high effect	extremely high effect	high effect	high effect	high effect
	increase	increase	increase	decrease	smoother	more compacted
Fiber shortening	unclear	unclear	high effect	unclear	moderate effect	Low effect
	unclear	unclear	increase	unclear	smoother	more compacted

Internal fibrillation increases fiber flexibility and fiber swelling caused by the process of delamination in fiber wall resulting in increasing in most of strength properties of paper through the effect of fiber segment activation. However, it has only small effect on internal bonding strength which is found to mainly rely on inter-fiber bonding. Moreover, internal fibrillation is found to have low impact on an optical, a surface and a structural property.

External fibrillation is involved in generation of fibrils resulting in enhancement of inter-fiber bonding which leads to the development of strength properties and smoothness of paper. Moreover, the growth in inter-fiber bonding also results in closely-compacted structure in paper network. However, the closely compacted structure notably decreases light scattering coefficient. The effect of external fibrillation on tensile strength contrast to the Hartman's study that external fibrillation has no effect on tensile strength (Hartman, 1985)

Fiber shortening is also found to affect various paper properties. Extremely very short fiber (fines) significantly develops tensile strength and stiffness. However, moderately short fiber does not show clear correlation on the strength properties. Therefore, the effect of fiber length on the tensile strength development is unclear. Nevertheless, shorter fiber tends to have smoother surface and denser structure.

Fiber curl and internal fibrillation were developed simaltaneously throughout the refining in both refiing methods so that is difficult to assess their relative importance in property development.

Recommendation

In this study, FSP was used to quantitatively estimate internal fibrillation based on previous study (Wang, 2006, Kang, 2007). Thermal porosimetry was planned to conduct to find out pore distribution of the fiber sample in order to confirm the correlation between FSP and internal fibrillation. However, the measurement was not successfully done because of the unreadiness of the measuring tools and limited time of the project.

Degree of external fibrillation and amount of fines were evaluated by Kajaani FS-200 fiber analyzer. Image analysis method was first planned to conduct to confirm the result. However, the analysis was only partially done because of the unreadiness of the software used in the analysis and limited time of the project.

Therefore, thermal porosimetry and further image analysis could be conducted to complete this study in the future study.

Chapter 5. Summary

The study found that Lampén refining, representing compressive refining, highly promoted external fibrillation, internal fibrillation, fiber straightening and somewhat shortens fibers. On the other hand, Voith refining, representing combination of abrasive and compressive refining, was found to highly promote external fibrillation, fiber straightening but moderately fiber shortening and internal fibrillation. Masuko refining, representing highly abrasive refining, strongly promoted both external fibrillation and fiber cutting while it only slightly developed internal fibrillation, zero span and fiber straightening. Furthermore, internal fibrillation was found to largely promote most of paper strength properties and to have a moderate impact on surface and structural properties while external fibrillation and fines greatly affected structural and surface properties.

References

1. Algar, W.H. and Giertz, W., Fiber structure and pulp properties. II. The influence of ultrasonic treatment on wood cellulose fibers. Svensk Papperstidn. 54(20):693-700(1951).
2. Berthold, J.,Salmen, L.: Inverse size exclusion chromatography for determining the relative pore size distribution of wood pulp. Holzforschung 51(1997):4,361-368.
3. Blomstedt, M., Asikainen, J., Lähdeniemi, A., Ylönen, T., Paltakari, J., and Hakala, T. K., 2010. Effect of xylanase treatment on dewatering properties of birch kraft pulp.Bioresources5(2),1164-1177. http://ojs.cnr.ncsu.edu/index.php/BioRes/article/view/BioRes_05_2_1164_Blomstedt_ALYPH_Xylanase_Dewatering_Poroperties/624
4. Borch, J., Lyne, M. B., Mark, R. E., Habeger, C. ed., 2001. Handbook of Physical Testing of Paper: Volume 2. New York: Marcel Dekker,Inc.
5. Brecht, W. and Siewert, W., 1996. Zur theoretisch-technischen Beurteilung des Mahlprozesses moderner Mahlmaschinen. - In: Das Papier. - 20(1966)1. - pp. 4-14
6. Brecht, W., Athanassoulas, M., and Siewaert H. W., 1965. Der Einfluss des Messerwinkels bei Hollandern and Refinern . W. fur Papierf. 19(4): 145-150.
7. Buchanan, J.G. and Lindsay, R.A., A note on structure of paper as revealed by the scanning electron microscope. Formation and Structure of Paper, Transactions of the 2nd Fundamental Research Symposium, Oxford, U.K., p. 101-108 (1961).
8. Clark J.D., Fibrillation, free water, and fiber bonding. Tappi J. 52(2):335-340 (1969)
9. Claudio-da-Silva, E., Marton, R. and Granzow, S., Effect of beating on wet web properties. Tappi J. 65(11):99-103 (1982).
10. Cohen, W.E., The performance and use of the Lampén mill in Australia. Proceedings of 2nd APPITA Annual Conference, Carlton, Australia, p.84-117 (1948).
11. Corson, S.R., Aspects of mechanical pulp fibre separation and development in a disc refiner. Pap. Puu. 71(7):801-814 (1989).
12. Cottrall, L.G., The influence of hemicelluloses in wood-pulp fibres on their papermaking properties. Pulp Pap. Mag. Can. 51(9):135-143 (1950).

13. Cress, O. and Bialkowsky, H., Some chemical and physical observations on hydration. Paper Trade J. 93(20):35-44(1931).
14. Dekker, J.: New insights in beating leading to innovative beating techniques. Proceedings of the 2003 PIRA Refining Conference, Stockholm.
15. Emerton, R.W., The specific external surface of fibre – some theoretical considerations. Pulp Pap. Mag. Can. 56(2):65-68 (1955).
16. Forgacs, O.L., The characterization of mechanical pulps. Pulp Pap. Mag. Can. 64:T89-T118 (1963).
17. Gallay, W., Some factors in the strength of paper. Tappi J. 32(10):457-462 (1949).
18. Gullichsen, J., Paulapuro, H., 2000. In-plane tensile properties. In: Gullichsen, J. and Paulapuro, H. ed., 2000. Papermaking Science and Technology book 16, Paper Physics. Otaniemi:Finnish Paper Engineers' Association/Paperi ja Puu Oy. Ch.4.
19. Harrison, A., The beating of wood pulp in laboratory and mill. Proceedings of the Technical Sections, Manchester, U.K., p.272-333 (1931).
20. Hartman, R.R., 1985. Mechanical treatment of pulp fibers for paper property development. Paper Raw Materials. Transactions of the 1985 Fundamental Research Symposium,BPBMA,Oxford, p.413-442.
21. Hiltunen E., 2000. Papermaking properties of pulp. In: Gullichsen, J. and Paulapuro, H. ed., 2000. Papermaking Science and Technology book 17, Paper Physics. Otaniemi:Finnish Paper Engineers' Association/Paperi ja Puu Oy. Ch.3
22. Hiltunen, E., 2003. On the beating of reinforcement pulp. Doctoral Thesis. Laboratory of Paper Technology, Reports Series A16. Helsinki University of Technology, Espoo.132 p.
23. Jayme, G. and Hunger, G., Electron microscope 2- and 3-dimensional classification of fibre bonding, Formation and Structure of Paper, Transactions of the 2nd Fundamental Research Symposium, Vol.1, Oxford, U.K., p.135-170 (1962).
24. Jayme, G. and Hunger, G., The fiber-to-fiber bonding in paper handsheets seen by means of electronmicrographs. Das Papier. 11:140-145 (1957).
25. Jayme, G.: Properties of wood celluloses. II. Determination and significance of water retention value. Tappi J.41(1958):11,180A-183A.

26. Kajanto, I., Niskanen, K. 2000. Dimensional stability. In: Gullichsen, J. and Paulapuro, H. ed., 2000. Papermaking Science and Technology book 16, Paper Physics. Otaniemi:Finnish Paper Engineers' Association/Paperi ja Puu Oy. Ch.7.
27. Kang, T. and Pualapuro H., 2007. New Mechanical Treatment fr Chemical Pulp, Journal of Preocess Mechanical Engineering. 220(2006)3,p.161-166.
28. Kang, T. and Pualapuro, H., 2006. Characterization of Chemical Pulp Fines. Tappi Journal. 5(2006)2, p.25-28
29. Kang, T. and Pualapuro, H., 2006. Effect of External Fribrillation on Paper strength. Pulp & Paper Canada. 107(2006),p.51-54
30. Kang, T., Somboon, P. and Pualapuro, H., 2006. Fibrillation of Mechanical Pulp Fiber. Paperi ja Puu. 88(2006)7, p.409-411.
31. Karrila, S., Raisanen, K.O., and Paulapuro, H., 1992. "Moving Belt Drainage Tester (MBDT)" Papermakers Conf. (Nashville, TN), Proc. (Book 1): 275-3OO,(TAPPI: April 5-8, 1992).
32. Kerekes, R. J., 1990. Characterization of pulp refiners by C-factor. Nord. Pulp and Pap. Res. J. 16(1): 3-8.
33. Kerekes, R.J. and Senger, J.J., 2006. Characterizing refining action in low consistency refiners by forces on fibres. J. Pulp Pap. Sci. 32(1):1-8(2006).
34. Kerekes, R.J, 2005. Characterizing refining action in PFI mills. Tappi J. 4(2005):3, 9−14.
35. Kerekes, R.J., 2005. Characterizing refining actions: linking the process to the refining results. Proceedings of the 2005 PIRA Refining Conference, Barcelona.
36. KnowPap, 2000. KnowPap Learning Environment for Papermaking and Automation. KnowPap -project 1997-2000. online availability at http://know.aalto.fi/pap/latest/english/knowpap_system/user_interfaces/frontpage.htm
37. Koskenhely, K., 2007. Refining of chemical pulp fibres. In: Paulapuro, H. ed., 2007. Papermaking Science and Technology book 8, Paper making part1, stock preparation and wet end. Otaniemi:Finnish Paper Engineers' Association/Paperi ja Puu Oy. Ch.4.
38. Kurhila, A., 2005. Developing a method for characterisation of fibre external fibrillation. Master Thesis, Helsinki University of Technology, Espoo,2005.

39. Laine C., Wang, X., Tenkanen, M., Varhimo, A., 2004, Change in the fiber wall during refining of bleached pine kraft pulp. Holzforschung 58(2004):3,233-240
40. Leider, P.J., and Nissan, A.H., 1997. Understanding the disc refiner: Measurement of treatment of fibres. Tappi J. 60(10):85-101 (1997).
41. Li, T.: Interaction between Water and Cellulose Fibres, Application of NMR Techinques. Doctoral Thesis, Royal Institute of Technology, Stockholm,1991
42. Lindqvist, H., Salminen, K., Kataja, J., Retulainen, E., Fardim, P.,and Sundberg, A, 2011. The Effect of Fines on Dewatering, Wet and Dry Web Properties. PaperCon 2011, P.887-894.
43. Lossada, A.A., Formento, J.C., Pino, A.L.: Refining selected actions on chemical pulps in a PFI beater. Part I: Effects of radial compression stresses. Paperi ja Puu 83(2001):4, 326−331.
44. Lumiainen, J. J., 1995. Specific surface load theory, In: 1995 PIRA 3rd International Refining Conference Proceedings, PIRA, Leatherhead, UK.
45. Lumiainen, J., 2000. Refining of chemical pulp fibres. In: Gullichsen, J. and Paulapuro, H. ed., 2000. Papermaking Science and Technology book 8, Paper making part1, stock preparation and wet end. Otaniemi:Finnish Paper Engineers' Association/Paperi ja Puu Oy. Ch.4.
46. Maloney, T.C., Paulapuro, H.: Thermoporosimetry of pulp fibers. Trans. Fund. Res. Symp. 2001, C.F. Baker, Ed., BPBMA, London,UK,p.897-926.
47. Maloney, T.C., Laine, J.E. and , Paulapuro, H., 1999. Comments on the measurement of cell wall water. Tappi J. 82(9):125-127(1999).
48. Mason, S.G., The specific surface of fibers - its measurement and application. Tappi J. 33(8):403-409 (1950).
49. Murphy, D.C.: Mechanical factors in beating. Appita J. 16(1962):1, 16−30.
50. Niskanen, K., Kajanto, I., Pakarinen, P., 2000. Paper Structure. In: Gullichsen, J. and Paulapuro, H. ed., 2000. Papermaking Science and Technology book 16, Paper Physics. Otaniemi:Finnish Paper Engineers' Association/Paperi ja Puu Oy. Ch.1.
51. Ora, M., Pitkäkuituisen mekaanisen massan jälkijauhatus, Master's thesis. Helsinki University of Technology, Department of Forest Product Technology, Espoo 1987, 190p.

http://www.pulpandpapercanada.com/paptac/PDFs/Mar04/MachineDesign.pdf

52. Page, D.H. and Sargent J.W., The fine structure of fibre bonding. Formation and Structure of Paper, Transactions of the 2nd Fundamental Research Symposium, Oxford, U.K., p.195-200 (1961)
53. Page, D.H., The beating of chemical pulps – the action and the effects. Papermaking Raw Materials, Transactions of the 9th Fundamental Research Symposium,Cambridge, U.K., p.1-37 (1989).
54. Page, D.H.: The mechanism of strength development of dried pulps by beating. Svensk Papperstidning 88(1985):3, R30−R35
55. Retulainen, E., Niskanen, K., Nilsen, N. (1998). Fibers and bonds. In: Niskanen, K.(ed.), Paper Physics, Papermaking Science and Technology Book 16, Fapet Oy, Jyväskylä, pp. 55-87.
56. SCAN-C 62:00, 2000. Water retention value. Scandinavian Pulp, Paper and Board testing committee.
57. SETH, R.S. The measurement and significance of fines. Pulp & Paper Canada 104(2): T47-T50 (February 2003).
58. Simpson, F.W. and Mason, S.G., Note on the treatment of cellulose fibres with ultrasonic waves. Pulp & Paper Canada. 51(8):70-72(1950).
59. Stone, J.E., Scallan, A.M. and Abrahamson, B., Influence of beating on cell wall swelling and internal fibrillation. Svensk Papperstidn. 19(10):687-694 (1968).
60. Stone, J.E., Scallan, A.M.: A structural model for the cell wall of water swollen wood pulp fibres based on their accessibility to macromolecule. Cellul. Chem. Technol. 2(1968):3,343-358
61. Strachan, J., Further notes on the hydration of cellulose in papermaking. Proceedings of the Technical Section, London, U.K., p.61-81 (1932)
62. Strachan, J., Some physical aspects of beating. Proceedings of the Technical Section, Manchester, U.K., p.171-194 (1938).
63. Strengell, K., Stenbacka, U., Ala-Nikkola, J., 2004. Advanced wire part simulation with a moving belt former and its applicability in scale-up on rotogravure printing paper. Pulp & Paper Canada 105(3): T62-66 (March, 2004).
64. Sundström, L., Brolin, A. and Hartler, N., Fibrillation and its importance for the properties of mechanical pulp fiber sheets. Nord. Pulp Pap. Res. J. 8(4):379-383 (1993).

65. Taniguchi, T. and Okamura, K., New films produced from microfibrillated naturalfibres. Polymer International. 47(3):291-294(1998).
66. Tasman, J.E., Pulp and Paper Manufacture Vol.II. Control, Secondary Fiber, Structural Board, Coating. Joint Textbook Committee of the Paper Industry. Macdonald, R.G. McGraw-Hill Book Company. New York, p.132-186 (1969).
67. Vainio, A., 2007. INTERFIBRE BONDING AND FIBRE SEGMENT ACTIVATION IN PAPER – OBSERVATIONS ON THE PHENOMENA AND THEIR INFLUENCE ON PAPER STRENGTH PROPERTIES. Doctoral Thesis. Laboratory of Paper Technology, Reports Series A29. Helsinki University of Technology, Espoo.
68. Wang, X., 2006. Improving the papermaking properties of kraft pulp by controlling horrification and internal fibrillation. Helsinki University of Technology, Espoo,2006
69. Wang, X., Maloney, T.C. and Pualapuro, H., 2003. Internal fibrillation in never dried and once-dried chemical pulp . Appita J.56(2003):6, 455-459.
70. Wang, X., Maloney, T.C. and Pualapuro, H., 2005. Chemical pulp refining for optimum combination of dewatering and tensile strength . Nord. Pulp Pap. Res. J. 20(2005):4, 442-447
71. Wang, X., Maloney, T.C. and Pualapuro, H., 2006. Fibre fibrillation and its impact on sheet properties. Paperi ja Puu.
72. Wang, X., Maloney, T.C. and Pualapuro, H., 2006. Improving the properties of never-dried chemical pulp by pressing before refining. Nord. Pulp Pap. Res. J. 21(2006):1, 135-139
73. Wathén, R., 2006. Studies on fiber strength and its effect on paper properties. Doctoral thesis, Helsinki University of Technology, Espoo, Finland.
74. Weise, U., Paulapuro, H., 1996. Relation between fiber shrinkage and hornification. Das Papier50(6), 328333 , 0031-1340

List of Appendices

Appendix 1: Data of measurement results

Appendix 2: Images of Sample from microscope

Appendix 3: Data of the result of pulp refined from Voith Refiner

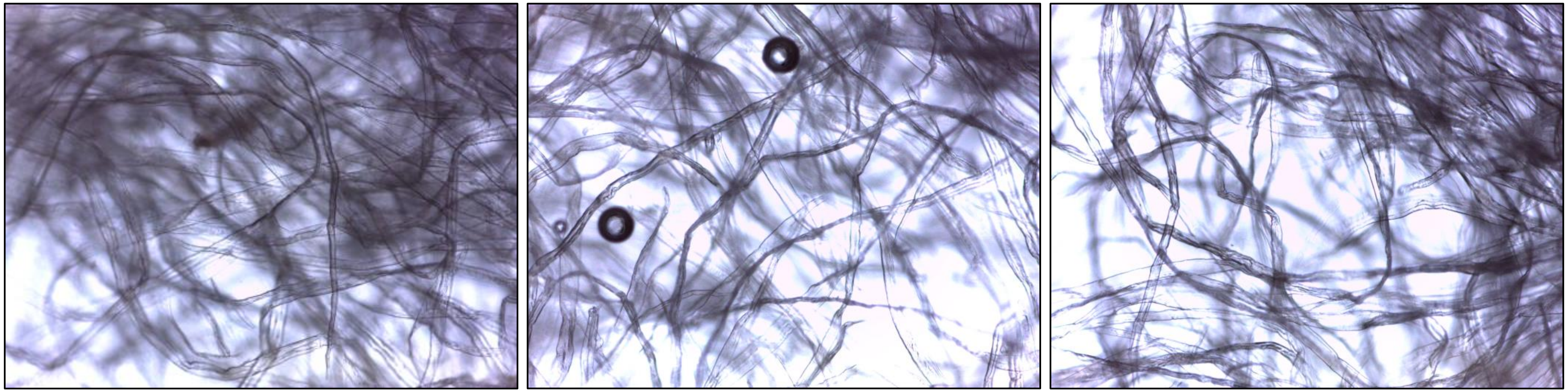
Appendix 1: Measurement results

	Specific Energy	fiber length (mm)	%fine	fibrillation (%)	fiber curl %	FSP (g/g)	SR	WRV (g/g)	Density	bulk (cm3/g)	Formation (SD)	Roughness	light scatter (m2/kg)	light absorption (m2/kg)
Lampén	0	2.19	4.27	0.66	18.45	0.47	21.00	0.57	0.40	2.48	4.90	1733.00	34.52	0.08
	3500	2.06	4.36	1.07	15.00	1.08	23.00	0.62	0.47	2.12	4.10	1529.00	32.24	0.10
	7000	1.89	5.02	1.60	14.30	1.17	29.00	0.66	0.61	1.64	3.90	1234.00	21.49	0.10
	11000	1.73	5.02	2.02	14.15	1.20	44.00	0.67	0.64	1.56	3.50	1093.00	18.69	0.11
Voith	0	2.26	2.94	0.70	19.40	0.68	16.00	0.59	0.43	2.34	5.09	1695.00	33.02	0.08
	75	2.28	3.38	0.91	17.80	0.87	18.00	0.59	0.49	2.06	5.20	1559.00	30.69	0.09
	150	2.28	3.40	1.47	16.95	1.02	26.00	0.61	0.55	1.83	6.00	1403.00	27.18	0.10
	225	2.15	3.86	2.11	16.90	1.08	41.00	0.62	0.60	1.66	5.70	1190.00	21.53	0.09
Masuko	1.5	1.67	9.02	2.48	17.5	-	81.50	0.57	0.74	1.36	-	1129.20	30.44	0.15
	0	-	-	-	-	-	11.50	-	-	-	-	-	-	-

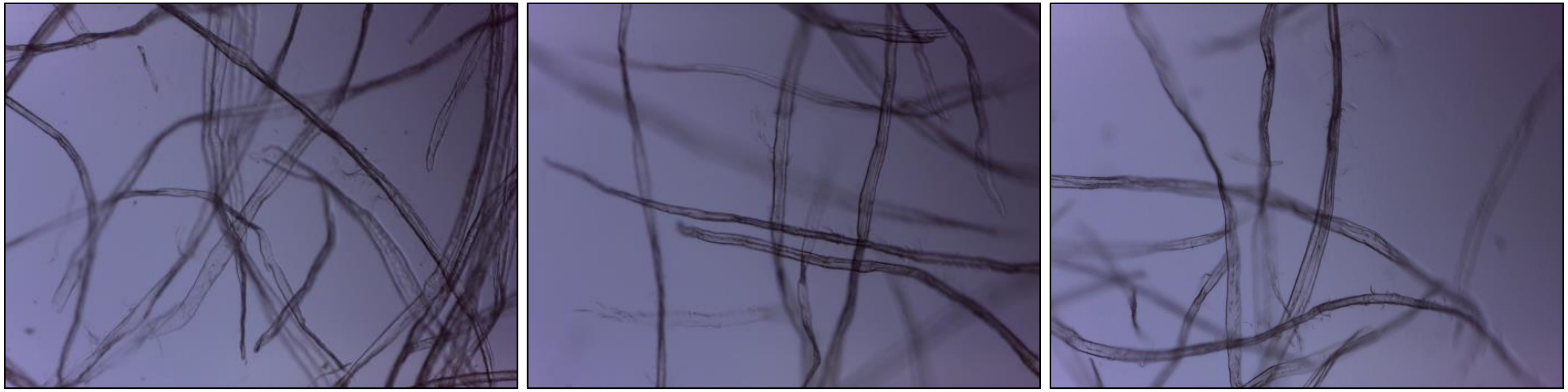
	ISO Brightness	opacity	Tensile index (kNm/kg)		Stiffness index (MNm/kg)		Stretch (%)		Zero span (kNm/kg)		internal bonding (J/m2)		MBF (%Dry matter content)		MBF (Retention)		Vacuum	Conductivity	PH
			Avg	**95% conf.**	**Avg**	**95% conf.**	**Avg**	**95% conf.**	**Avg**	**95% conf.**	**Avg**	**95% conf.**	**Avg**	**95% conf.**	**Avg**	**95% conf.**			
Lampén	87.25	76.08	11.05	0.68	1.64	0.09	1.67	0.32	117.7	0.25	52.10	5.54	19.90	1.26	92.60	8.09	29.21	12.5	7.24
	86.05	71.44	30.07	1.38	3.70	0.23	2.69	0.28	146.3	0.52	113.90	6.14	19.21	1.24	88.70	5.74	30.89	11.1	7.88
	83.75	63.96	79.56	3.13	7.06	0.26	3.61	0.12	156.9	0.43	489.70	7.45	19.10	0.39	81.00	7.48	33.95	10.95	7.61
	82.58	60.58	89.36	3.48	7.61	0.25	3.85	0.26	167.0	0.42	583.30	25.67	19.21	0.14	82.40	0.86	35.76	8.5	7.5
Voith	87.24	75.08	20.68	1.04	2.63	0.13	3.10	0.35	120.6	0.39	99.60	6.09	20.48	0.30	97.20	2.79	32.92	7.5	7.5
	86.27	71.47	43.11	2.68	4.70	0.24	3.95	0.20	136.8	0.33	212.10	3.38	18.63	0.27	96.30	5.69	34.35	4.1	7.66
	85.05	69.70	62.78	0.23	5.57	0.23	4.37	0.34	136.31	0.93	386.20	12.98	16.47	1.29	98.10	4.13	36.01	4.5	7.96
	84.57	66.90	74.47	4.09	6.47	0.25	4.51	0.19	151.9	0.50	496.00	14.33	16.03	0.68	98.20	2.88	37.49	4.85	7.8
Masuko	83.78	69.56	56.6	3.59	5.511	0.12	5.68	0.59	120.9	0.18	714.75	-	-	-	-	-	-	13.02	7.89
	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-

Appendix 2: Images of Sample from microscope

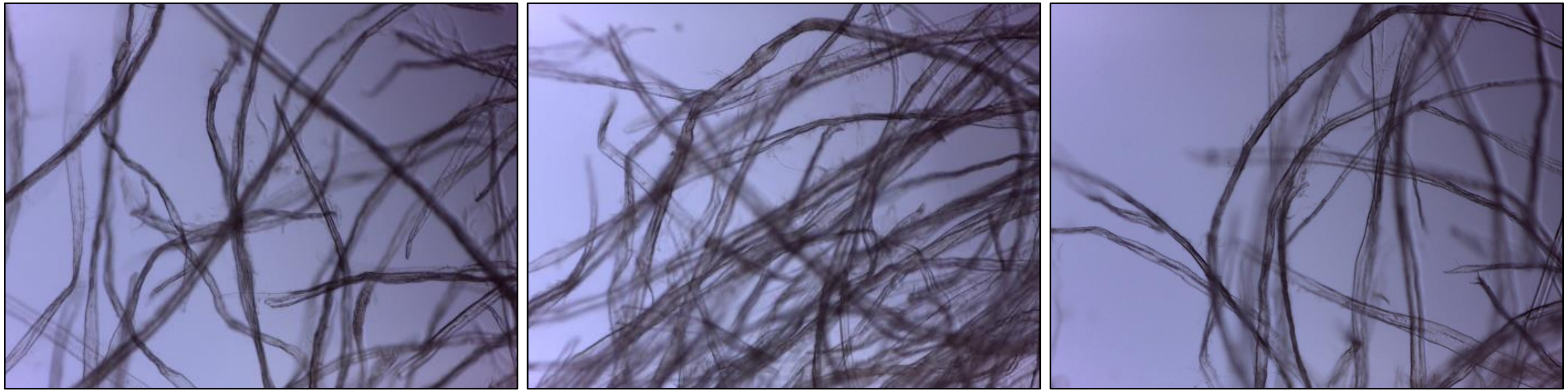
0 kWh/t of Voith-refined pulp



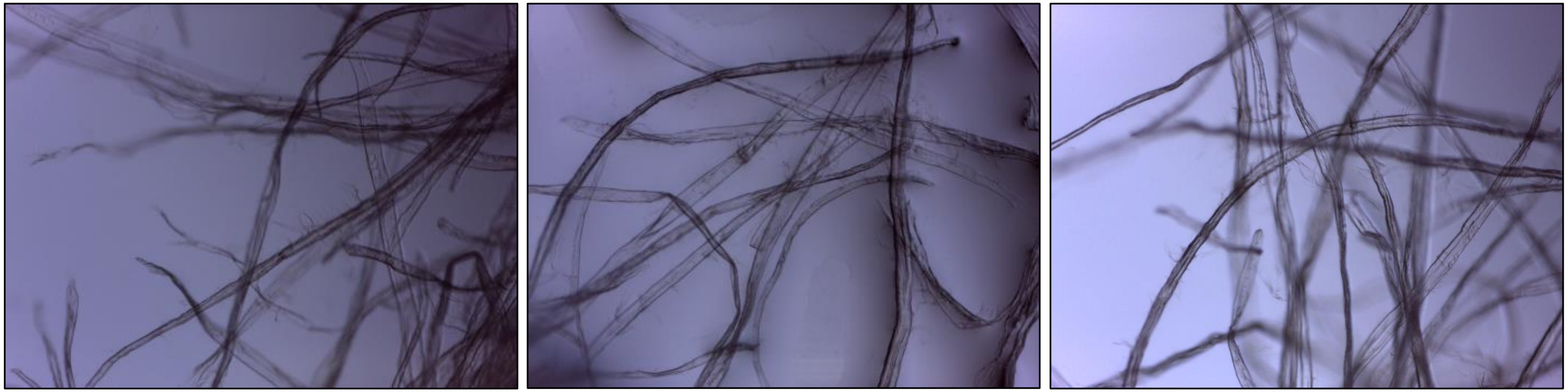
75 kWh/t of Voith-refined pulp



150 kWh/t of Voith-refined pulp

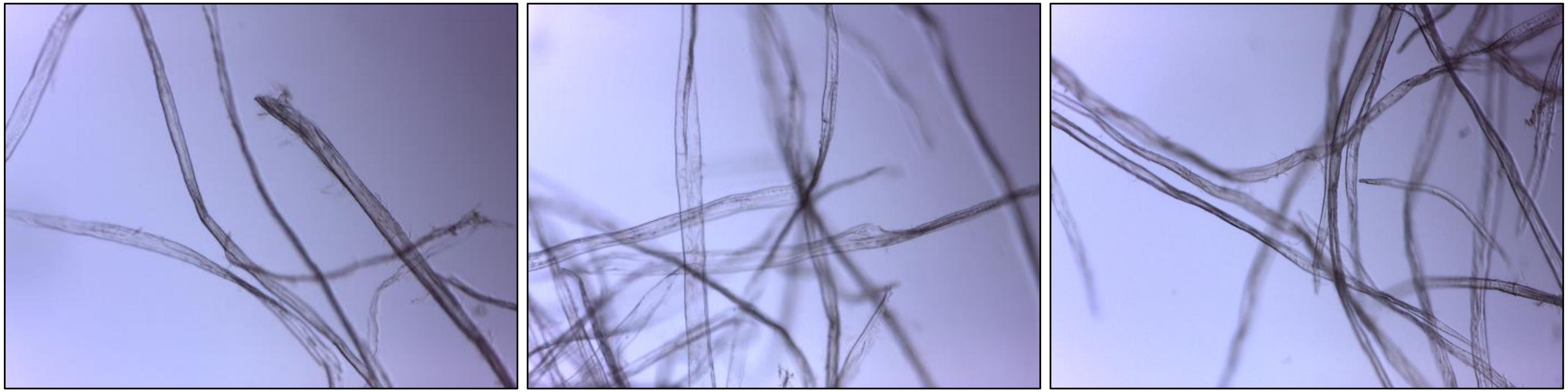


225 kWh/t of Voith-refined pulp

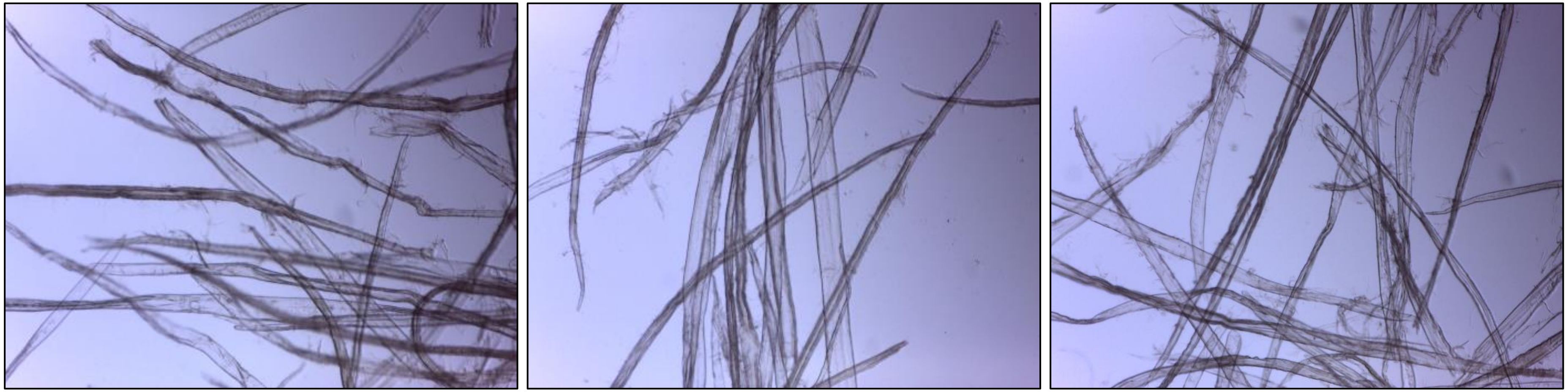


0 revolution of Lampén-beaten pulp

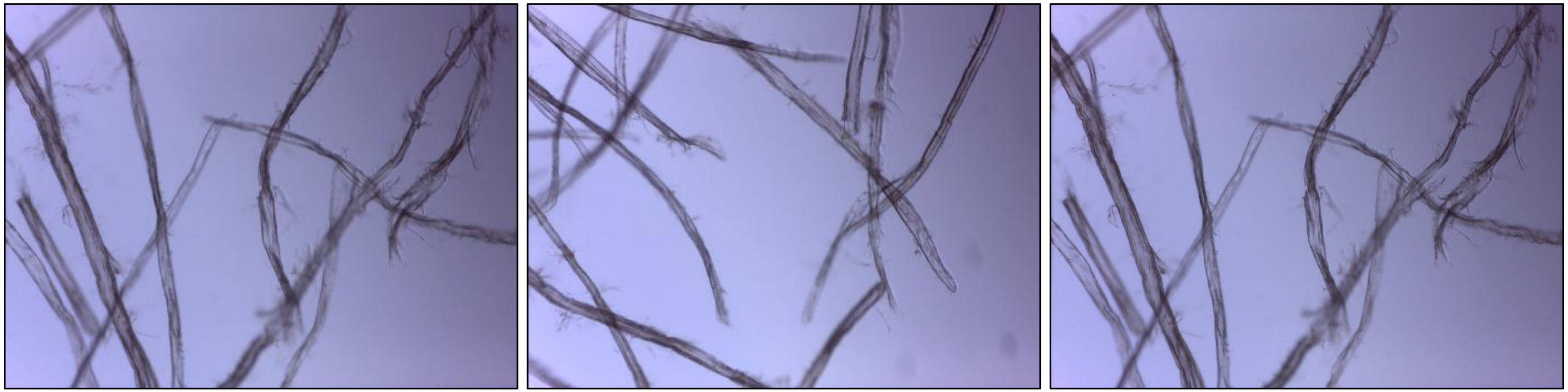
3000 revolutions of Lampén-beaten pulp



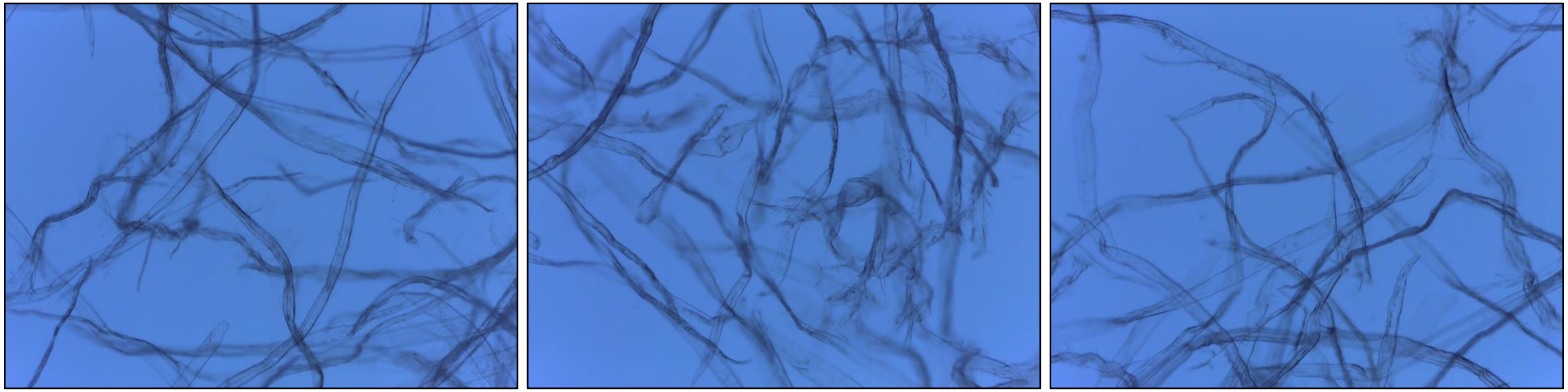
7000 revolutions of Lampén-beaten pulp



11000 revolutions of Lampén-beaten pulp



1.5 kWh/t of Masuko-refined pulp



Appendix 3: Raw data of the parameter and result Voith Refining used in the experiment

VOITH PAPER | **Laboratory Refiner LR 40** | **Trial Protocol**

Date:	2013.12.13 07:47:49	Furnish:	voith refining	Spec. edge load [J/m]: SEL	2.50
Trial no.:	468	Fillings designation:	3-1,0-60	CEL [km/s]:	0.67

Refiner speed [rpm]:	2000	Susp. volume [l]:	37.5	Pulp quantity:		Pulping time [min]:	10
No-load power [kW]:	2.03	Consistency setp. [%]:	4.00	air dry [g]:	37500	Dwelling time [min]:	10
with water "0", stock "1":	0	Consistency av. [%]:	4.00	oven dry [g]:	1500	Density [g/cm3]:	1.00

Sample no.:	1	2	3	4	5	6
Refining time [s]	0	233	481	696	0	0
Sample wet mass [g]	1540	1710	1550	1620	0	0
Sample oven dry mass [g]	62	68	62	65	0	0
Remaining mass [g]	1438	1370	1308	1243	1243	1243
Inlet pressure [bar]	1.0	0.7	0.7	0.7	0.0	0.0
Total load power [kW]	1.80	3.51	3.68	3.69	0.00	0.00
Net refining power [kW]	0.00	1.47	1.65	1.65	0.00	0.00
Total spec. energy [kWh/t]	0	158	343	511	511	511
Net spec. energy [kWh/t]	0	66	149	224	224	224
Rotor position [mm]	0.000	21.916	21.986	22.023	0.000	0.000
Temperature [°C]	26.6	30.2	34.1	37.7	0.0	0.0
Consistency [%]	4.00	4.00	4.00	4.00	0.00	0.00
SR- value [SR]	0.0	233.3	481.0	695.9	695.9	695.9
SEL [J/m]	0.0	2.2	2.5	2.5	0.0	0.0
MEL [J/m]	0.0	9.6	10.7	10.8	0.0	0.0
Reserve3 [X_3]	0.0	233.3	481.0	695.9	695.9	695.9